ISOTOPES AND RADIATION TECHNOLOGY

A Quarterly Technical Progress Review

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FEATURE ARTICLE:

Mass Production and Practical Applications of Actinide Elements

By Glenn T. Seaborg

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for ISOTOPES INFORMATION

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Isotopes and Radiation Technology

Vol. 6, No. 1 Fall 1968

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Isotopes and Radiation Technology

is a Technical Progress Review prepared by the Isotopes Information Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development
Isotope Technology Development
Food Irradiation
Process Radiation Development
Source Development
Isotopic Heat and Power Development
Applications in Life Sciences
Safety
Isotopes Around the World
Miscellaneous

P. S. Baker, Robert H. Lafferty, Jr., Joan E. Carden, Martha Gerrard, R. E. Greene, F. E McKinney, and F. J. Miller, *Editors*
A. F. Rupp, *Adviser*

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Isotopes and Radiation Technology
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Nuclear Safety
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Reactor and Fuel-Processing Technology
Argonne National Laboratory

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Other U. S. Government agency reports identified in this journal generally are available from CFSTI.

Private-organization reports should be requested from the originator.

United Kingdom Atomic Energy Authority (UKAEA) and Atomic Energy of Canada Limited (AECL) reports are available at USAEC depository libraries. UKAEA reports are sold by Her Majesty's Stationery Office, London; AECL reports are sold by the Scientific Document Distribution Office, Atomic Energy of Canada Limited, Chalk River, Ontario, Canada. UKAEA and AECL reports issued after March 1, 1967, are sold by CFSTI to purchasers in the United States and its territories.

FEATURE ARTICLE

Mass Production and Practical Applications of Actinide Elements*

By Glenn T. Seaborg†

I am indeed honored to be the introductory speaker at this 5-day symposium on macroscopic studies of the actinides. Since almost my entire scientific career has been devoted to the actinides and since I spent most of that career in this geographic area, it is a great temptation to be nostalgic and reminiscent about the "good old days." However, nostalgia has had its place during the several anniversaries we have celebrated recently commemorating such events as the first weighing of plutonium and the first sustained nuclear fission reaction.

Research on the actinides remains one of the most dynamic areas of scientific endeavor. Programs under way for the large-scale production of numerous isotopes of the synthetic actinide elements, together with unexpected practical applications of a number of these isotopes, point to an exciting future for the manufacture and exploitation of these elements. The magnitude of these possibilities is an aspect of the transuranium field that was not foreseen even as recently as a few years ago. I shall attempt in this discussion to describe and forecast some of these unexpected and intriguing possibilities.

A modern version of the periodic table (Fig. 1) is helpful in explaining the status of actinide synthesis. Each of the 11 synthetic actinide elements (atomic numbers 93 to 103, inclusive) was discovered as the result of its first production and identification in tracer quantities. In the case of the last three, mendelevium (number 101), nobelium (number 102), and lawrencium (number 103), the discovery experiments were performed with almost incredibly small amounts—essentially a single atom at a time. This will continue to be the case for the elements beyond lawrencium, i.e., the "transactinide" elements.

Very exciting—at least to me—has been the recent experimental work on the tracer chemistry of the elements mendelevium, nobelium, and lawrencium in aqueous solution. This type of work had not been possible previously with nobelium and lawrencium. A remarkable series of experiments has been performed with these elements using them on a "one atom at a time" basis in chemical reactions. E. K. Hulet, R. W. Lougheed, J. D. Brady, R. E. Stone, and M. S. Coops at Livermore, and J. Maly and B. B. Cunningham at Berkeley, have observed and studied the (II) as well as the (III) oxidation state of mendelevium. A. Ghiorso, J. Maly, T. Sikkeland, and R. J. Silva at Berkeley have found a very stable (II) oxidation state, as well as a quite unstable (III) oxidation state, of nobelium. And perhaps most exciting of all, A. Ghiorso, M. Nurmia, T. Sikkeland, and R. J. Silva have found, in experiments at Berkeley, that lawrencium exhibits a stable (III) oxidation state. In the absence of any known longer lived isotopes of these elements, they managed to determine these definitive chemical characteristics by using as tracers the 3-min ^{255}No and the 35-sec ^{256}Lr! These results are particularly gratifying to me because, in the case of all three of these elements, they support predictions based on the actinide concept. Thus we can feel confident that predictions being made for the chemical properties of the transactinide elements now stand on a firmer base.

The discovery of the synthetic actinide elements was followed, for the elements with atomic numbers 93 to 99, inclusive, by their isolation in macroscopic (weighable) quantities. The first isolation of

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†Chairman of the U. S. Atomic Energy Commission.

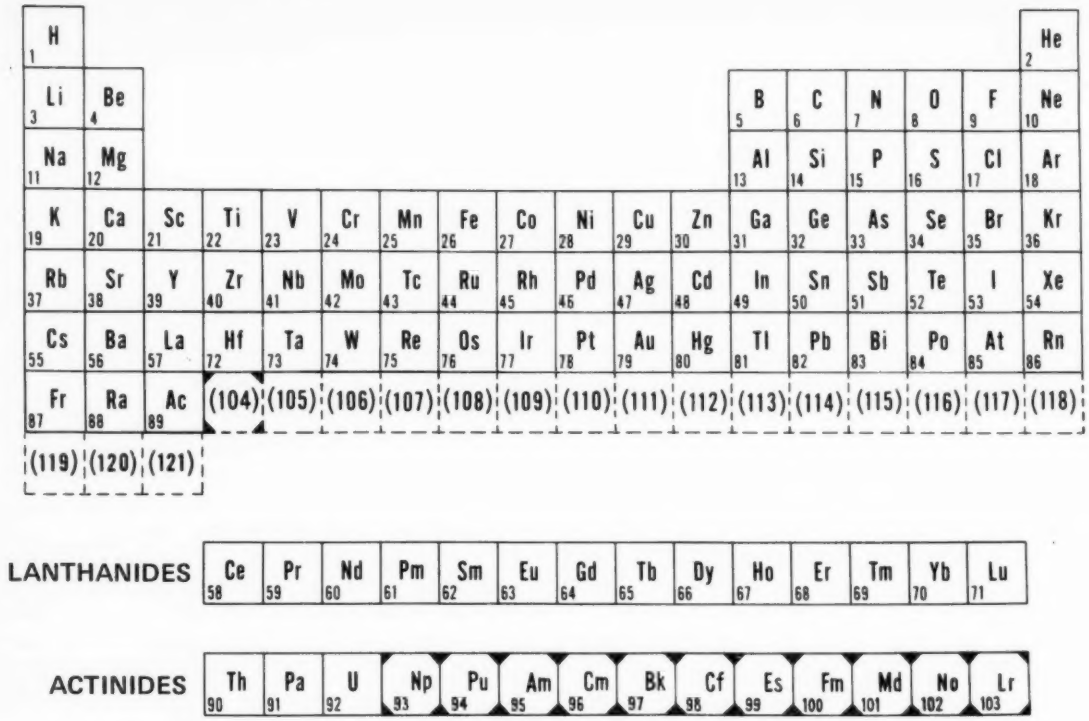

Fig. 1 Modern periodic table of the elements.

einsteinium (element 99) in large enough quantities to be visible was accomplished just recently by Cunningham and coworkers at Berkeley. In each case this phase of the element's history occurred a number of years after its discovery. And following this winning of the element in macroscopic quantities, it has been obtained in ever increasing amounts. Plutonium, of course, has been produced in the largest quantity by far, while both the production and the prospects for the production of the transplutonium elements decrease in a general way with increasing atomic number.

Although the half-lives of the isotopes of these elements suggest that einsteinium is the highest one that will be available in very useful quantities, it is interesting to note that the recent discovery of relatively long-lived ^{257}Fm (half-life, 95 days) and ^{258}Md (half-life, 54 days) may make it possible to extend the work with macroscopic quantities to elements 100 and 101. This does not seem to be a likely possibility for transmendelevium elements (numbers beyond 101).

This discussion will not be concerned with the applications and production of ^{239}Pu, which has such well-known utility as a source of energy in nuclear weapons and nuclear power reactors. It will be concerned, rather, with (1) the practical applications of a number of other transuranium isotopes and (2) their production potentialities.

Applications of the Actinides

With respect to the practical applications of the actinides, there is much more to be expected from work with them than simply increasing fundamental knowledge, as important as this is. Among the numerous practical applications that can be visualized in the near- and long-term future are the use of ^{238}Pu and ^{244}Cm as energy sources in long-lived, compact power units; ^{238}Pu to power medical prosthetic devices such as cardiac pacemakers or even artificial hearts; ^{241}Am to provide radiation for industrial measurement of thickness, density, and distance; and ^{252}Cf as a portable neutron point source in neutron

radiography, neutron activation analysis, hydrology, and medical therapy.

The Actinides as Energy Sources

An excellent indicator of the increased availability of actinide elements is their application in energy-conversion systems as sources of thermal power. This technology is only a little more than 10 years old, and it is much less than that if we measure only the period of time from the first practical application.

If an alpha-particle-emitting isotope is considered as a heat source, we are making use only of the 5 Mev or so of kinetic energy possessed by each helium ion liberated from the parent in the decay process, as compared with 40 times this energy liberated in the fission of a heavy atom of roughly the same mass number. Early workers in nuclear physics were inclined to dismiss the potential for nuclear energy because they did not know about fission, and the energy release rate of an alpha- or beta-particle emitter did not appear to be too attractive, considering the effort required to obtain from nature macroscopic quantities of such isotopes as ^{210}Po or ^{226}Ra.

With the advent of the nuclear reactor and our consequent ability to manufacture heavy actinide elements by transmutation from the more stable lighter actinide elements of nature, we now find that there is real merit in considering how to apply their unusual properties to the solution of real problems.

Consider for a moment a typical transuranium alpha-particle emitter such as ^{238}Pu, whose production I shall describe a little later. Its alpha decay half-life of about 90 years is equivalent to an initial energy-emission rate of about a half a watt per gram of pure isotope. Now if the decay is integrated over a 10-year period, assuming this to be a reasonable useful life for its application as a heat source, we find that 1 g of this isotope has liberated about 42 kw-hr of energy (Fig. 2). To accomplish this same release of energy by the combustion of a hydrocarbon, we would have had to consume 3300 g of butane, for example, and would have had to supply 12,000 g of oxygen to enable the combustion to proceed. Thus there is a weight ratio of at least 15,000 between the nuclear and conventional fuels, and this would be much larger if my calculations were based on air rather than oxygen and did not assume exact stoichiometry.

Obviously, then, one is not going to use radioisotope heat sources to supplant the butane torch of the home workshop or the electric hot plate or range in the kitchen. Radioisotopes are still expensive compared to chemical fuels. In the example I just used, 1 g of ^{238}Pu can be taken to be worth about $1000 (a cost that will certainly drop in the future). The combined cost of its thermal equivalent in compressed butane and oxygen would be of the order of $10 for the 10-year use period. Of course, the plutonium isotope is not being used very effectively from an economic viewpoint in this example, because only about 7% of its potential alpha-particle decay energy would be expended in that first 10-year period, and I have not given any dollar credit to the undecayed fraction remaining. If I prorate the cost accordingly, then the isotopic heat source costs only about $70.

The most effective comparison, however, is the relative size of these two energy sources. The 1 g of

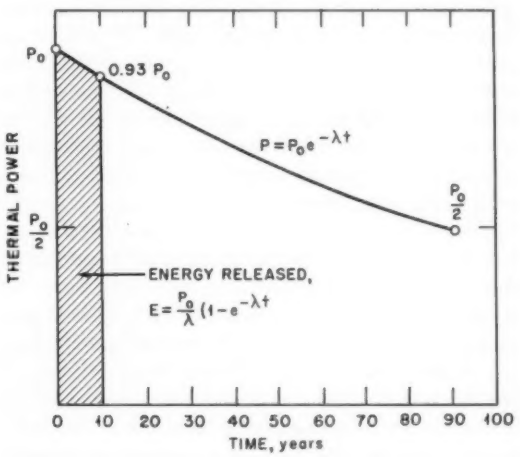

Fig. 2 Energy release from ^{238}Pu.

^{238}Pu, in the form of the oxide, occupies only about one-tenth of a cubic centimeter, while it would be necessary to cart around at least two very large compressed-gas cylinders to supply the butane and oxygen.

Energy-Conversion Systems. Weight, volume, and cost are not the only factors that determine if a radioisotope heat source is practical. Very few requirements have appeared so far for a supply of thermal energy per se. Most applications need electric power or mechanical power, and thus a means for converting the thermal power of the radioisotope to the other types of power must be provided; this leads to further technical and economic complexities. The development of energy-conversion systems for

radioisotope heat sources has been the responsibility of the AEC, although other federal agencies have participated heavily also—particularly in prototype testing. Some private development of isotopic power supplies has also occurred in recent years.

Three general types of conversion systems have been given the most attention: dynamic, thermoelectric, and thermionic.

1. Dynamic Energy Conversion. Under "dynamic systems" I include schemes that make use of thermodynamic cycles, such as the Rankine, the Brayton, and the Stirling (Fig. 3*a*). Engines operating on these cycles have the characteristic of requiring the repetitive use in a closed system of a heated fluid which may or may not change phase during the cycle. Working fluids that have been considered include water, mercury, and inert gases, depending on the cycle. Their output is mechanical power, but by coupling them to a rotating generator we can obtain electric power readily. These cycles are all in common usage in systems where fossil fuels provide the source of energy, but in this case the working fluid is often employed only once in a so-called "open" cycle. Conversion efficiency is determined by the temperature extremes of the cycle and, thermodynamically, can range as high as 50% or so, although this figure would be difficult to achieve in practice.

2. Thermoelectric Conversion. Thermoelectric conversion (Fig. 3*b*) is a more recent innovation and makes use of the Seebeck effect in doped semiconductors. It has the distinct advantage of requiring no moving parts to convert heat directly to electricity, but it, too, has a thermodynamic limitation to conversion efficiency, with actual efficiencies ranging from 5 to 10%, depending on the semiconductors used and the temperature extremes achieved.

3. Thermionic Conversion. Thermionic conversion, the principle used in the vacuum diode in which a heated filament emits sufficient electrons to provide a useful current and voltage, also can be employed with thermal-energy sources derived from radioisotopes (Fig. 3*c*). No moving parts are involved, and the efficiencies attainable are projected to be about twice those now available from thermoelectric conversion, i.e., 10 to 20%.

Development of Practical Systems. All three modes of radioisotope energy conversion face the same general development problem: in order to maximize the conversion efficiency and hence reduce the inventory of radioisotope required for a given power output, the heat source and conversion equipment must op-

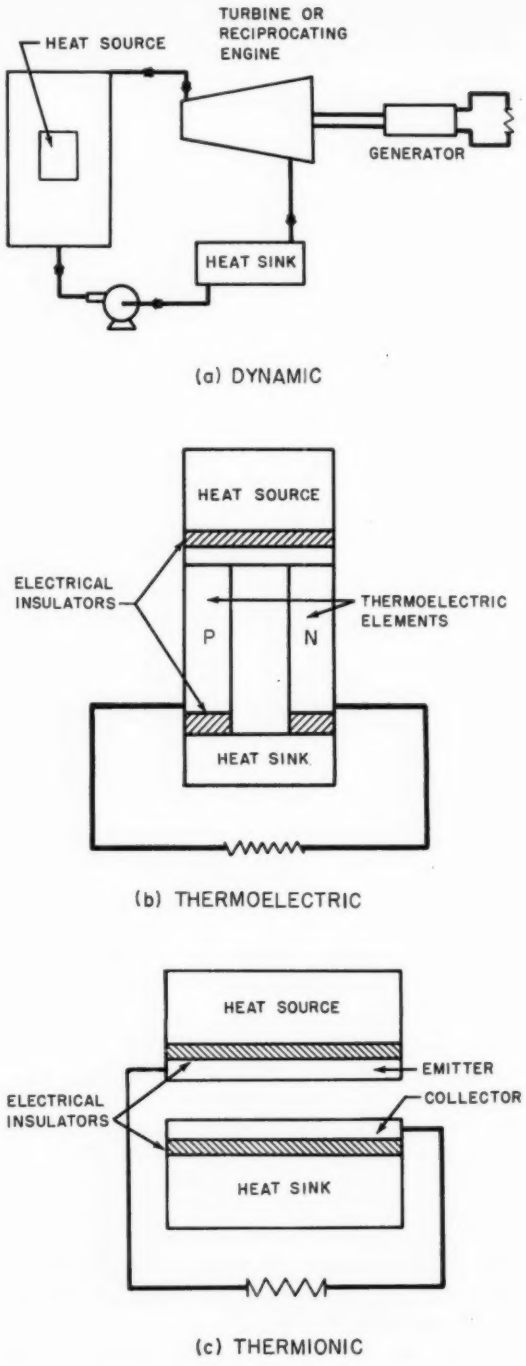

Fig. 3 Energy-conversion systems.

erate at the highest possible temperature. As chemists, you will not be surprised that most of the effort involved in developing radioisotope power supplies has been concerned with inventing, modifying, and testing high-temperature materials, particularly with regard to the chemical form of the radioisotope, its encapsulant, and the materials for construction of the conversion equipment.

The development effort on radioisotope power supplies, now running at about $35 million per year in the AEC's budget, can be justified only because there are near-term and long-range applications for these devices which cannot be met by batteries or other chemical conversion processes. What are some of these applications which we find to be so exciting technically and so important as to warrant the investment being made? Let me categorize them by the nature of the energy requirement.

1. Heat. First, there are indeed some applications that need only thermal energy. Consider the case of a space satellite or probe containing an electronics package which has to operate in an environment whose temperature approaches absolute zero but which functions optimally at a higher temperature. Perhaps the spacecraft is on a mission away from the sun where the incident solar energy is insufficient to warm it, or has landed on the moon, where there is no solar heating for 14 earth-days at a time. Some thermal energy is available from resistive heating of the electronic components, but this may be insufficient. If solar cells are the source of electric power for the space vehicle, it is almost immoral to go to the expense and technical difficulty of making electric power and then to degrade it to thermal power for the sole purpose of providing additional heating for the spacecraft. On the other hand, a small radioisotope heat source can provide the compensating thermal energy with 100% reliability and suffers only from the fact that it cannot be turned on and off. In a manned spacecraft a continuous and reliable source of heat may be required for long-duration missions—not only for temperature conditioning but for such purposes as water recycle and recovery. A joint AEC–Air Force development program for an isotopically heated water-recovery system is well along the development path and looks very attractive in its economic and technical aspects (Fig. 4).

Another purely thermal application is more down-to-earth. You may have seen some publicity about the joint program of the AEC and the Navy to develop a heater for the suits of deep-sea divers and aquanauts. The first prototype has been completed and is under test (Fig. 5). The heating unit consists of 750 g of ^{238}Pu, which produce 420 watts of thermal power to heat a fluid that circulates through the "veins" of the suit. A development problem still to be solved in this application is reduction of the ionizing radiation level from the heat source. Although ^{238}Pu is primarily an alpha-particle emitter, it also decays by spontaneous fission in a small fraction of the total decay events, and gamma radiation is emitted as part of both the alpha and spontaneous-fission decay processes. The background level of gamma rays and the energetic neutrons from fission cannot be ignored. Other contributors to the radiation level are neutrons resulting from (α,n) reactions on light-element impurities in the chemical form of the isotope and gamma radiation from the decay of ^{236}Pu, a nuclear impurity.

2. Electric. The next category of use for radioisotope heat sources is the electric power supply. I am quite sure that you are all familiar by this time with one or more of the so-called SNAP (Systems for Nuclear Auxiliary Power) devices that have already been used in every environment of the earth. Some of these power supplies use fission products as heat sources, primarily because they are less expensive than the heavy-element alpha-particle emitters. All devices in actual use employ the thermoelectric conversion principle. The SNAP device's versatility is well demonstrated by its ability to function at the bottom of the ocean, in the frigid climates of the Arctic and Antarctic, and in outer space.

I will mention specifically here only three new radioisotope power supplies which are in the final stages of development and which have exciting futures ahead of them. SNAP-19, a 50-watt supply consisting of twin thermoelectric generators fueled with ^{238}Pu will be launched aboard the Nimbus B satellite later in 1968 on a mission to collect data on the earth's weather conditions (Fig. 6). SNAP-27 is also a thermoelectric power supply fueled with ^{238}Pu, but of much different design than used heretofore. It is an integral part of the scientific equipment to be left on the moon by the Apollo astronauts (Fig. 7). One of the novel features associated with it is that the heat source will be transported to the moon in a container separate from the conversion apparatus and will be inserted in the converter after the landing on the moon. This approach was deemed necessary to provide additional assurance that the integrity of the heat source would be maintained if it became necessary to abort the mission during any part of the sequence from earth launch to lunar landing.

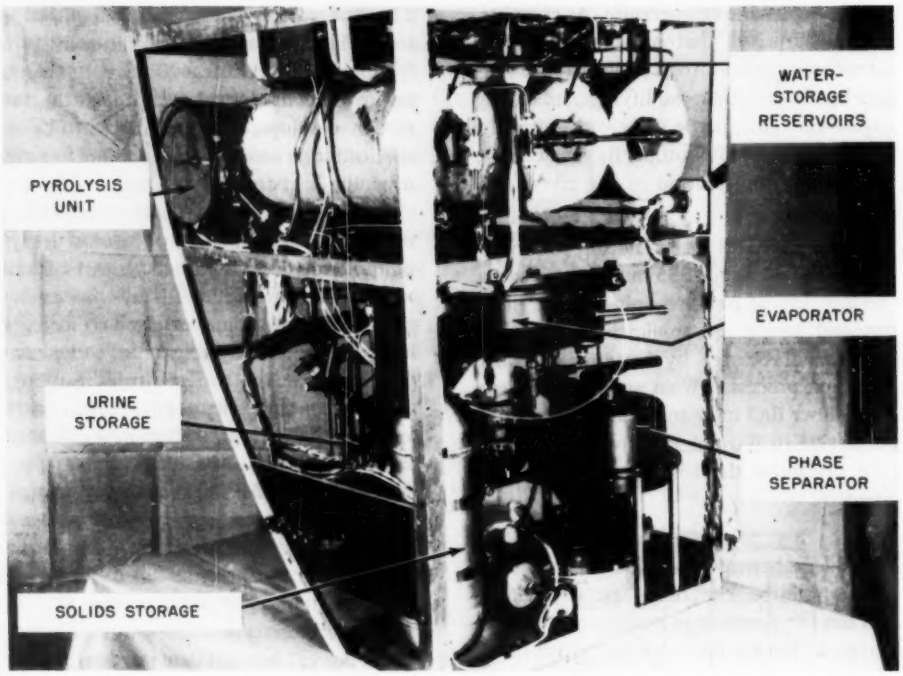

Fig. 4 Water-recovery system for astronauts (^{238}Pu).

Fig. 5 Radioisotope swimsuit heater.

In this age of miniaturization, it is not too surprising that a tiny isotopic power supply is also under development. Its field of application is not so esoteric as that of space exploration, but it may be of far more immediate importance to us. I am referring to the device now in the testing phase which is designed to power a cardiac pacemaker (Fig. 8). As you know, several forms of heart disease are characterized by failure of the heart to receive a large enough or regular enough electrical stimulus to cause normal pulsation. Stokes-Adams disease is a significant one of these heart malfunctions. Correction is achieved by implanting an artificial electrical stimulator or pacemaker in the thoracic or abdominal cavity and attaching electrodes to the heart surface. Conventional pacemakers use chemical batteries as their source of energy, and the batteries limit the life of the devices to from 2 to 3 years. The isotopic power supply, which would replace the implanted chemical batteries, is designed to last at least 10 years and should therefore reduce the frequency of replacement of the power supply for the pacemaker. Of course, the radioisotope heat source would last much longer than 10 years, but we are not so sure about the electronics or the patient! The

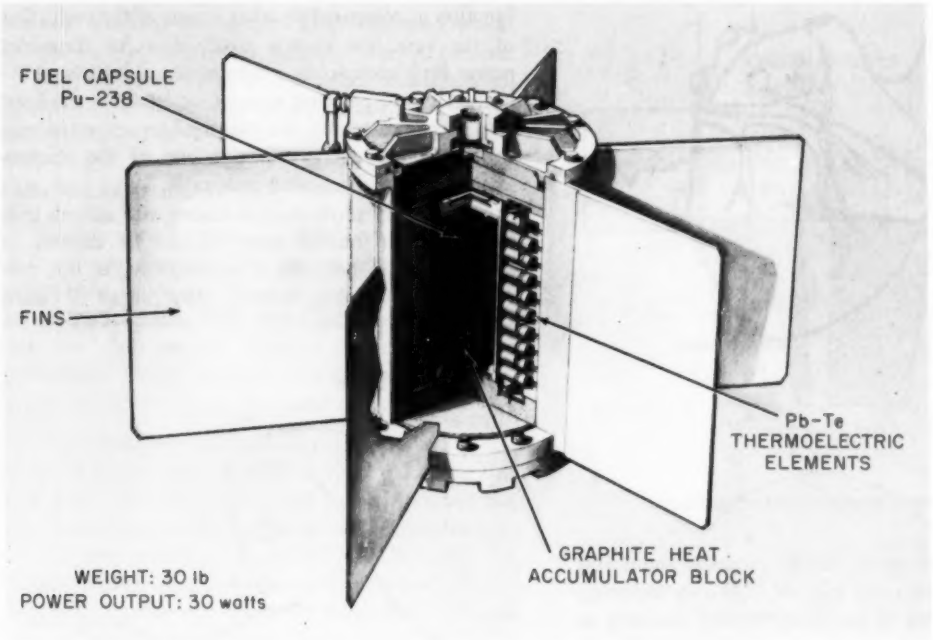

Fig. 6 SNAP-19 power supply for Nimbus B satellite.

Fig. 7 SNAP-27 nuclear power for Apollo lunar surface experiment package.

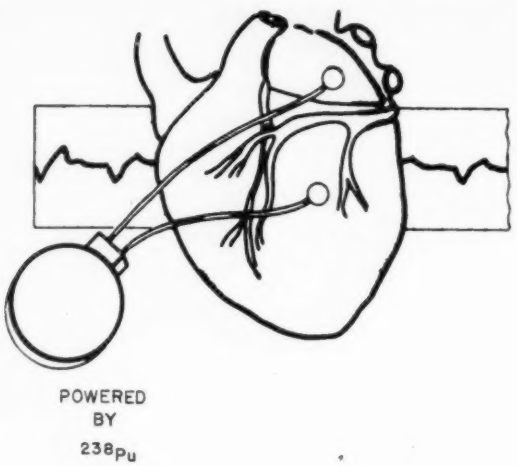

Fig. 8 Radioisotope-power cardiac pacemaker.

nuclear pacemaker power supply produces about 160 μw of electric power and will use ^{238}Pu as its energy source. Only 300 mg of isotope are needed. As many as 10 thousand pacemakers per year may be required.

3. Mechanical. The next category of utility that comes to mind is based on the mechanical power supply. One example will suffice here, and it constitutes one of the most intriguing potential applications for nuclear energy that has yet been devised. Transplantation of human hearts from one individual to another has received worldwide attention recently, and the results obtained have been discussed in every home with access to a newspaper or radio. The public is less well acquainted with the modest program being carried out by the National Heart Institute of the National Institutes of Health to develop mechanical devices to assist the pumping action of a failing human heart or—in the extreme—to completely replace the human heart.

The ultimate objective of this program is to produce an artificial heart that, after implantation within the patient's body, would restore the person to a condition where he would be a useful member of society again. This means, of course, that the artificial organ would have to have a compact and reliable power supply—no wires plugged into the light socket permitted!

Problems associated with the development of an artificial heart are so complex and severe that I cannot give full assurance that such a device will be available in any predictable time period; yet there is reason for optimism because the heart is relatively simple in its function as compared to other organs of the body. One of the very few known possibilities for furnishing power for a completely implantable artificial heart lies in the radioisotope heat source (Fig. 9). Since the heart is a mechanical pump, the heat need be converted only to mechanical energy through one of the thermodynamic cycles mentioned previously.

Preliminary physiological studies with animals indicate that the human body should be capable of dissipating the heat that, unavoidably, is not converted to pumping action. Only about 7 watts

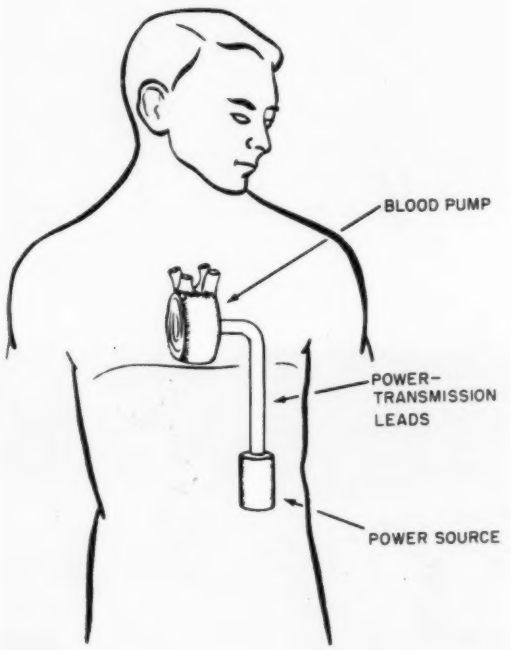

Fig. 9 Artificial heart components.

(maximum) of pumping power are required to sustain the life of a person in normal activity, and this pumping power drops to 2 or 3 watts under quiescent conditions, as in sleeping. If an implanted thermodynamic engine and pump can be made to operate at 20% overall efficiency, then between, say, 15 and 35 watts of thermal power would be required. Should the radioisotope power supply prove to be the best, and if all the other developmental problems can be overcome,

demands for radioisotopes to meet the need would amount to requirements for tons per year! At present, ^{238}Pu, properly purified to reduce its radiation level, appears to be the leading candidate for the artificial heart power source.

Radiation Applications

Turning now from the consideration of isotopes as heat sources, let us give our attention to those applications which are based on the radiation properties of the actinides. Each of the actinides has isotopes that are alpha-particle emitters, which decay by spontaneous fission and hence are sources of neutrons, or that produce characteristic gamma rays or both.

Americium-241. Starting with the lower end of the atomic number range, ^{241}Am is finding a variety of uses. It is in plentiful supply, and even larger quantities will be available in future years, as will be pointed out. Its 433-year half-life minimizes the necessity for frequent decay correction but, unfortunately, makes its specific activity too low to be considered at present as an alpha-particle heat source.

One property of particular interest in the decay of ^{241}Am is the 60-kev gamma ray emitted in about 35% of the alpha-particle decays. This predominant photon emission is excellent for use in various types of measuring equipment such as thickness and level gages because its attenuation coefficient in materials of moderate-to-low density is neither too high nor too low.

Another use for the gamma ray from ^{241}Am takes advantage of its ability to excite elements in the middle of the periodic table in order to detect their presence and determine their relative amounts by observing their characteristic X rays produced by the fluorescence process. An unusual application of this technique, now being studied by Dr. Paul Hoffer at the Argonne Cancer Research Hospital, involves moving a collimated 300-mc ^{241}Am gamma source and a solid-state radiation detector around the area adjacent to a patient's thyroid gland (Fig. 10). The excited stable ^{127}I present in the thyroid emits X rays at an intensity proportional to the amount and distribution of iodine, and an indication is thereby obtained of the functional adequacy of the thyroid. This method could conceivably replace or supplement the now widely used radioiodine uptake and scanning technique because there is no residual radioactivity left in the patient, and the radiation dose to the patient is markedly reduced.

For periods of time up to a few years, the correction for decay of ^{241}Am is often less than the statistical counting error and can be ignored. Popular applications for ^{241}Am include its use in static eliminators, smoke detectors, and radiography units.

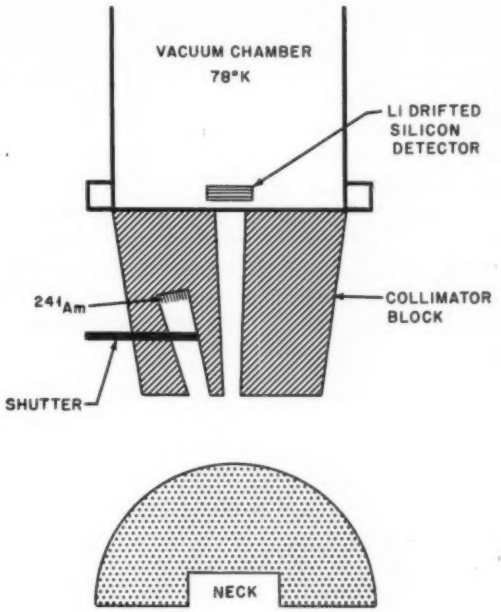

Fig. 10 Apparatus for thyroid diagnosis.

Americium-241 as an alpha emitter is important in oil-well logging, where it functions as an Am–Be neutron source.

Other Actinides. Several other actinide elements have alpha-particle-emitting isotopes that are available in large enough quantity to be of commercial or scientific interest. The isotopes ^{239}Pu, ^{238}Pu, ^{242}Cm, and—surprisingly—^{254}Es have been used as alpha-particle sources for one or more of the following purposes:

1. To produce neutrons by the (α,n) reaction with elements of low atomic number such as beryllium.
2. To provide external beams of alpha particles as the excitation medium in new methods of chemical analysis, such as alpha-excited X-ray analysis and alpha scattering analysis.
3. To provide a source of alpha particles for methods of analysis of physical properties such as density or thickness based on absorption or back-scattering of alpha particles.

Whereas 15 or 20 years ago I could note with wonder the emerging industrial uses of isotopes, I must now turn to more exotic applications to avoid reciting

the commonplace. It is with some personal pride that I note that the first use of radioisotopes on an extraterrestrial body—the moon—involved the isotopes of two actinide elements, ^{242}Cm and ^{254}Es. The 162-day ^{242}Cm was the source of alpha particles in the equipment on board Surveyor V, which performed the first direct chemical analysis of the lunar surface by alpha-particle scattering and alpha–proton reaction techniques. The source strength was about 100 mc. The ^{254}Es was placed near the alpha-particle detectors to act as an energy marker at 6.44 Mev. The maximum energy of the ^{242}Cm alpha particles is 6.11 Mev.

Future Applications of the Actinides: ^{252}Cf. As far as future applications of the actinides as radiation sources are concerned, I look forward with the greatest interest to the fruition of developments now taking place with ^{252}Cf. Since this isotope will be available in at least gram quantities within a few years, it is not at all out of place to speculate on its large-scale uses, all of which at present are based on the fact that this isotope, with its nominal 2.6-year half-life, decays by spontaneous fission in about 3% of the decay events. Since spontaneous fission is accompanied by neutron emission, this is equivalent to a neutron yield of about 4.4×10^9 n/sec per curie of ^{252}Cf—about 300 times as many neutrons per alpha-particle disintegration as can be obtained with the best (α,n) neutron source (e.g., a Ra–Be source). One curie of ^{252}Cf weighs about 2 mg, so it is just about as intense a neutron source as a power reactor! The general advantage of ^{252}Cf over machine or reactor sources of neutrons is its inherent compactness and portability. Among the applications being studied are:

1. Radiotherapy. Neutron irradiation of cancerous tissue is a promising approach in cancer therapy and is important because neutrons overcome the radiation resistance of anoxic cells in tumors. Several applicators in the form of needles, each containing a few micrograms of californium, have already been supplied to medical researchers. The convenience and economy of the needles are most evident here since patients would not have to be taken to reactor or accelerator sites for treatment. This is a good example of the mountain coming to Mohammed!

2. Radiography. Neutron radiography is a technique complementary to the more familiar gamma or X radiography. It is expected to be of value in the medical or biological fields because it offers improved delineation of soft-tissue morphology and elimination of excessive bone contrast. It may also be useful in nondestructive inspection methods where large thicknesses of heavy materials must be penetrated by radiation. High-atomic-number materials have relatively low mass-absorption coefficients for neutrons. Californium-252 neutron sources for this purpose would be much more adaptable to field or laboratory use than reactors or accelerators. A problem still to be resolved, however, is the development of a good imaging technique for neutrons of fission energy. Most of the development work on imaging with neutrons has been done in the thermal region.

3. Activation Analysis. Since neutron activation analysis is now a widely accepted technique, the application of ^{252}Cf as a portable neutron source is quite apparent. It would be particularly advantageous for mineral exploration and in remote regions, such as the ocean floor or the surfaces of other planetary bodies.

4. Radiation Effects. A number of early experiments come to mind in which unshielded reactors have been operated above ground for the purpose of generating sufficient neutrons to determine their effect on the surrounding ecology or to measure the attenuation afforded by various types of shielding. This kind of research is important for the future, primarily for defense research purposes. It has been proposed that ^{252}Cf be used instead of a reactor or accelerator neutron source. A 40-g californium source would produce 10^{14} n/sec and would require very little in the way of supporting facilities other than means to remove the 1.6 kw of thermal power that would be generated.

5. Hydrology, Petroleum Exploration, and Other Industrial Applications. Almost all oil wells and an increasing number of water wells are logged for geophysical purposes by one or more techniques, including neutron scattering. Alpha–n neutron sources are in common use for this purpose, (in fact, this is one of the largest uses for ^{241}Am), but ^{252}Cf would offer the advantages of greater neutron intensity and much more compact size relative to other isotopic neutron sources or to small accelerators which produce neutrons by the D–T reaction. Other possibilities for its use include the routine measurement by neutron scattering of the water content of soils, wood, concrete, coal, and other bulk materials, and on-line neutron activation analysis of flowing process streams.

Reactor Fuels Beyond ^{239}Pu

Although I am not discussing here the well-known applications of ^{239}Pu as a nuclear fuel, some possible

special applications of heavier actinide isotopes for this purpose are worth mentioning briefly.

The potential importance of the fission properties of higher mass-number isotopes of plutonium and of the heavier actinide elements is not often considered. For example, the availability of increasing amounts of americium and curium isotopes may make feasible the construction of thermal-neutron reactors fueled with such isotopes as the 152-year ^{242}Am, with a thermal-neutron fission cross section of about 6000 barns, or the 9300-year ^{245}Cm, with a cross section of about 2000 barns; and, perhaps further in the future, we may consider the use of such isotopes as the 360-year ^{249}Cf and the 800-year ^{251}Cf, which also have been reported to have very high thermal-neutron fission cross sections.

With such isotopes as these, it may become feasible to construct a very small reactor with a minimum amount of fuel which someday might find importance.

Perhaps closer at hand is the possibility of using ^{239}Pu and heavier plutonium isotopes in "Phoenix fuel," an application that has been studied in the past at such installations as Pacific Northwest Laboratory. Here advantage is taken of appropriate combinations of these isotopes, so that one of the isotopes serves as a nonfissionable burnable poison whose neutron-capture product is fissionable. Mixtures of ^{239}Pu, nonfissionable ^{240}Pu, and fissionable ^{241}Pu (the Phoenix fuel) make possible the design of a reactor system that, from the nuclear standpoint, is able to sustain almost constant reactivity over very long periods of time. It is conceivable that in future reactor applications this may prove to have some very important advantages.

Actinides for the Chemist

The actinide elements constitute a fascinating transition series of great intrinsic scientific interest to the chemist. In addition, as these elements become a more familiar and important part of our technical and economic life, it is natural to expect that chemists will want to study their chemical and metallurgical properties in greater depth than is now being done. Such exploration of the actinides will be more convenient and fruitful as some of their especially long-lived isotopes become available in macroscopic quantities (as the result of intensive neutron irradiations, followed by electromagnetic separation) which are nearly isotopically pure. Problems arising from radiolysis of solutions, degradation of crystal structures, handling of highly radioactive materials, and transmutation by decay will thus be minimized.

The prospects for this approach appear to be very good:

- Neptunium-237 already qualifies in this regard.
- Plutonium-244, with its 76-million-year half-life, will be a relative pleasure to work with in the laboratory; its preparation in macroscopic quantity will require electromagnetic separation from other plutonium isotopes.
- Americium-243 has a half-life almost 20 times as long as ^{241}Am, is more convenient for chemical research, and will be available in large quantities.
- By comparison with ^{242}Cm and ^{244}Cm, the isotopes ^{245}Cm through ^{248}Cm, singly or in mixtures, would almost be considered by the radiochemist to be stable! Curium-247, with a half-life of 16 million years, is the best single isotope of curium. The 470-thousand-year ^{248}Cm is also good and is easier to prepare in high isotopic purity (e.g., as the alpha-particle-decay product of ^{252}Cf). These heavier curium isotopes will also require magnetic separation for high isotopic purity.
- Berkelium-247 has a half-life of about 1400 years, but no easy way to produce it in macroscopic amounts has yet appeared.
- The 360-year ^{249}Cf can, fortunately, be prepared in isotopically pure form by chemically isolating its beta-particle-emitting parent, ^{249}Bk, and letting it decay to the daughter ^{249}Cf. The even longer lived ^{251}Cf (half-life about 800 years) would require very complete electromagnetic separation from "hot" ^{252}Cf in order for it to be useful for chemical studies. Beyond this point the state of affairs promises to remain "hot" or scarce.

Large-Scale Production of Actinides

With respect to the production of the actinides, the present indications are that these other isotopes can be and will be produced in quantities that are amazing, at least to those who recall their birth in terms of only a few atoms. We are talking about quantities measured in tons for two of these isotopes, ^{238}Pu and ^{244}Cm, and in kilograms or hundreds of grams for heavier isotopes up to californium.

If it appears at times that my optimism is beginning to carry me away, let me note right here that the chemistry of plutonium was initially studied with picogram quantities of this element as a tracer, that the first weighing of the element involved a few micrograms, and that we are now predicting without many qualms the production within a few years of plutonium

in power reactors in quantities of hundreds of metric tons. The scale-up in quantities thus amounts to about 10^{20}, a number more meaningful to astrophysicists than to chemists!

Starting with the isotope ^{239}Pu, the sequence of thermal- or resonance-neutron absorption in nuclear reactors leads to such typical transmutation products as ^{242}Pu, ^{243}Am, and ^{244}Cm. Plutonium-242 can be produced in quantity by special irradiations of ^{239}Pu in large reactors such as the AEC's production reactors, and this method has been used up to now to obtain the target material for successive irradiations. The future extensive operation of nuclear power reactors will produce isotopes such as ^{243}Am and ^{244}Cm in massive quantities, and amounts of other higher isotopes that diminish in quantity with increasing mass number will be formed by neutron irradiation in other reactors. Figure 11 shows the main line of buildup of these isotopes. A projection of the quantities of these isotopes which will be available in future years is given in Table 1.

What are the present and anticipated capabilities for producing the actinide elements in large quantities in the United States? To answer this question, I will essentially exclude consideration of accelerators and charged-particle reactions for this purpose because the quantities produced are usually too small and too expensive for practical applications. I also exclude, therefore, production of elements above fermium, since they have not yet been observed as the product of neutron-induced nuclear reactions.

Neptunium-237 is an important isotope because it is the precursor of ^{238}Pu. Fortunately, it is produced incidentally in nuclear reactors by the $(n,2n)$ reaction on ^{238}U and by successive (n,γ) reactions on ^{235}U and ^{236}U to form ^{237}U, which undergoes beta decay to ^{237}Np (Fig. 12). This long-lived alpha-particle emitter (half-life of 2 million years) is now being recovered from irradiated AEC production-reactor fuels. At least two private companies are considering recovering it from spent power-reactor fuels, and if our predictions of the rate of growth of the nuclear power industry materialize, it can be expected that 200 kg/year from this source could be available by 1975 and about 500 kg/year by 1980.

Plutonium-238 is produced by the thermal-neutron irradiation of ^{237}Np in AEC production reactors (Fig. 12). Its production rate has been limited by the amount of ^{237}Np available. However, essentially all requirements for it are being met. The AEC has had a commitment to produce 900 kg of ^{238}Pu for use by the National Aeronautics and Space Administration (NASA) in the 1964–1980 time period, contingent upon materialization of NASA long-range plans for a variety of space missions. Sufficient production-reactor space and neptunium target material should be available for this purpose. In fact, this isotope could be available in ton quantities to meet other large requirements that might develop, including the artificial heart and pacemaker programs. As I indicated previously, it would be desirable to produce a higher-quality isotope–that is, with reduced ^{236}Pu content for biological applications. This could be accomplished by storing the ^{238}Pu while the relatively short-lived (2.85 years) ^{236}Pu decayed preferentially. A 10-fold reduction in ^{236}Pu content occurs in about 10 years. Such a procedure would be relatively expensive and would require another chemical separation of the decay products at the end of the storage period. A more attractive approach would be to minimize the formation of ^{236}Pu, which is produced by the $(n,2n)$ reaction on ^{237}Np, by increasing the proportion of thermal neutrons in the reactor used to convert ^{237}Np to ^{238}Pu.

Americium-241 is valuable for its own uses and as a starting material for isotopes of higher mass number. It also is produced incidentally as the beta-particle-decay product of the ^{241}Pu which is present in all plutonium formed in production and power reactors. The AEC has been the only domestic source of this isotope in years past. It will be available in much larger quantities from increased power-reactor operation in the future, particularly because power reactors subject the plutonium formed to higher neutron exposures than production reactors, thereby increasing the relative concentration of ^{241}Pu. However, this isotope would be mixed with ^{243}Am at the time of discharge of the spent fuel. To recover pure ^{241}Am, it would be necessary to separate the mixture of plutonium isotopes in the conventional manner and allow the ^{241}Am to grow in as the result of decay of ^{241}Pu. If plutonium is being well utilized in plutonium recycle reactors or fast breeder reactors, it might not be economically attractive to store the plutonium, to allow the decay to occur, and then to process the plutonium again to recover the americium.

It is with some satisfaction that I note the current and future plans for the recovery of large quantities of ^{237}Np and ^{241}Am as by-products of nuclear reactor operation, a course of action that I have advocated with various degrees of insistence for more than 20 years.

Curium isotopes begin to cause special production problems. Curium-242, produced by thermal-neutron

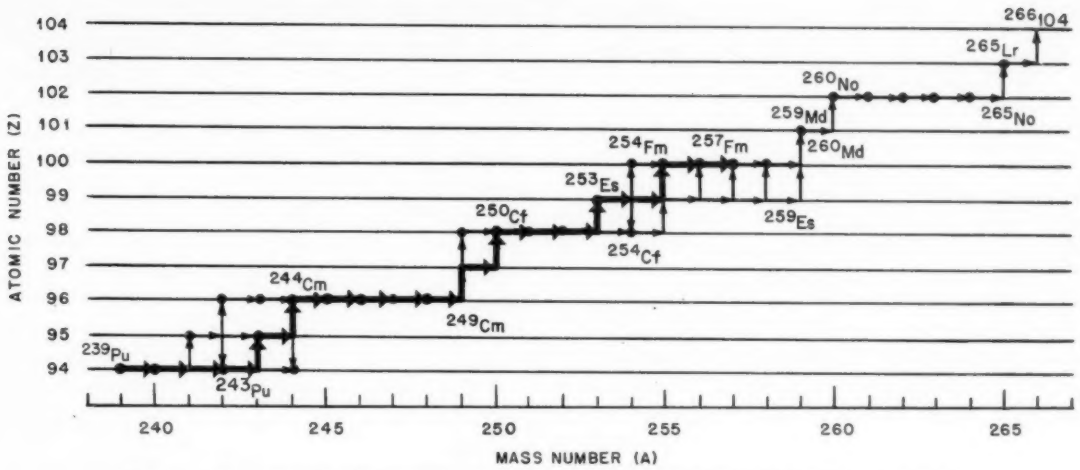

Fig. 11 Nuclear reactions for the production of heavy elements by intensive slow-neutron irradiation.

Table 1 Estimated Availability of Separated Transuranium Elements in the United States by 1975 and 1980

Element	Atomic No.	Principal isotope	Annual production rate, g	
			1975	1980
Neptunium *	93	^{237}Np	3×10^5	1×10^6
Plutonium *	94	^{239}Pu	1×10^7†	3×10^7†
Americium *	95	$^{241,243}Am$	8×10^4	3×10^5
Curium *	96	^{244}Cm	2×10^3‡	1×10^4‡
Berkelium §	97	^{249}Bk	10^{-1}	
Californium §	98	^{252}Cf	1¶	
Einsteinium §	99	^{253}Es	10^{-2}	
		^{254}Es	10^{-5}	
		^{255}Es	10^{-6}	
Fermium §	100	^{257}Fm	10^{-11}	

*From U. S. civilian power reactors only.
†Includes higher-mass-number isotopes.
‡Includes small quantity of higher-mass-number isotopes.
§From HFIR only.
¶Excludes lower-mass-number isotopes that are present.

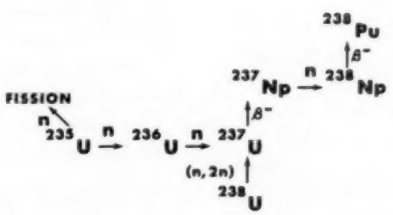

Fig. 12 Production of ^{238}Pu from ^{235}U and ^{238}U.

irradiation of ^{241}Am, has a relatively short half-life and is formed in high yield only if, on a comparative basis, the neutron flux is high and the irradiation time short. Although ^{242}Cm has been considered as a heat-source candidate in applications where the mission life is relatively short–perhaps up to 6 months–and where use could be made of its intense thermal-emission rate–120 watts/g–no such application has reached fruition. However, hundreds of grams per year of ^{242}Cm could be produced on relatively short notice if the need arose.

Curium-244, the alpha-particle-emitting isotope with an 18-year half-life, is proposed as an alternative to ^{238}Pu in heat sources. It has the advantage of exhibiting a higher thermal emission rate than the latter isotope and the potential of production earlier in larger quantities, but it suffers the disadvantage of having a higher radiation level due to neutrons from spontaneous fission.

Six kilograms of ^{244}Cm are now in the final stages of production at Savannah River; the process involves irradiating ^{239}Pu for several years, since five successive neutron captures are involved. To accomplish even the irradiation within this time scale, it has been necessary to increase the thermal-neutron flux of a reactor substantially above levels found in other reactors. The highest continuous peak neutron flux ever reported, 6.1×10^{15} n/(cm^2)(sec), has been achieved as part of the heavy-element program at Savannah River. Studies indicate that this can be increased still further to 2.5×10^{16}, which would correspond to a time-average flux at reactor center of 0.7×10^{16}. In the future, recovery of Am–Cm fractions from spent power-reactor fuels could provide excellent starting material for production of ^{244}Cm in subsequent high-flux irradiations. Costs would be reduced substantially by this approach, which would make feasible the production of tons of this isotope.

The High Flux Isotope Reactor

As we go to higher mass-number curium isotopes or beyond the curium isotopes, conventional power or production reactors no longer serve to provide the intensive neutron fluxes required to increase the mass number and atomic number. At least three different means appear to be available for this purpose.

First, we have the high-performance research reactor epitomized by the High Flux Isotope Reactor, or HFIR, at Oak Ridge National Laboratory. In operation since 1966, this 100-Mw reactor can produce a flux as high as 5×10^{15} n/(cm^2)(sec) which is perturbed to about half this value when a ^{242}Pu target of 250 g is inserted in the flux-trap region of the core. This reactor has now produced about 7 mg of ^{252}Cf, which have been isolated in the transuranium processing facility (TRU) adjacent to the reactor. The ^{242}Pu target material comes from Savannah River, where it is prepared by high-flux neutron irradiation of ^{239}Pu. By the mid-1970's, the HFIR will be producing ^{252}Cf at the rate of 1 g/year, along with 0.1 g of ^{249}Bk (and therefore potentially of its useful daughter, ^{249}Cf), 10 mg of ^{253}Es, 0.03 mg of ^{254}Es, and lesser amounts of fermium isotopes.

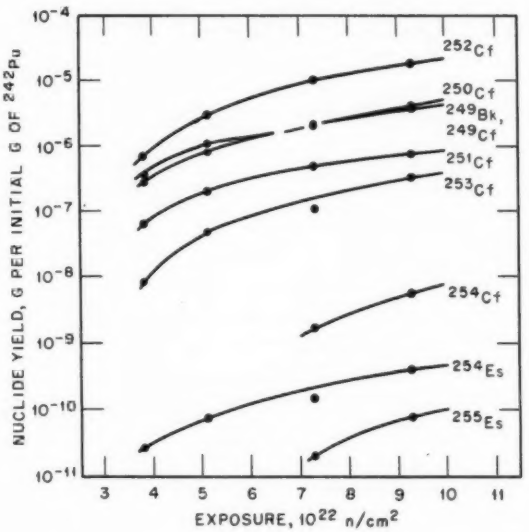

Fig. 13 Yield of transcurium isotopes resulting from high-flux neutron irradiation of ^{242}Pu in Savannah River Reactors.

The Savannah River production reactors, operating in the thermal high-flux mode, also produce these higher isotopes, of course, as indicated in Fig. 13. Twenty-five milligrams of ^{252}Cf have been produced there as part of the ^{244}Cm campaign described previously. If a Savannah River reactor were to operate at a high thermal flux for 2 years, an additional 3 to 4 g of ^{252}Cf could be available by about 1973.

Yields of actinide isotopes that can be anticipated from reactors like HFIR are shown in Fig. 14.

The Resonance Reactor Concept

Another possibility for increasing the production rate of heavy elements involves changing the mode of operation of a Savannah River reactor to produce a

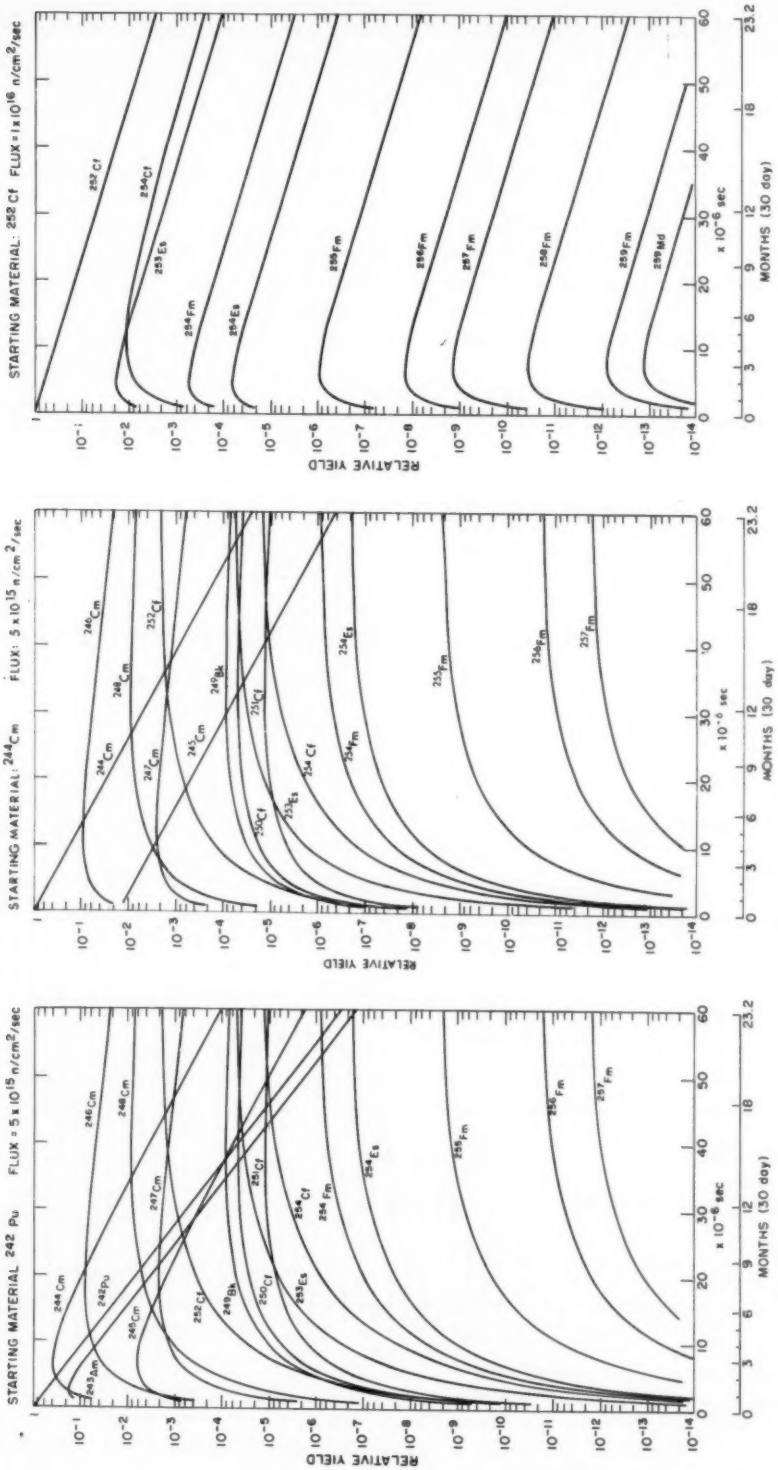
STARTING MATERIAL: 252 Cf FLUX = 1 x 10^16 n/cm^2/sec
STARTING MATERIAL: 244 Cm FLUX: 5 x 10^15 n/cm^2/sec
STARTING MATERIAL 242 Pu FLUX = 5 x 10^15 n/cm^2/sec
RELATIVE YIELD
MONTHS (30 day)
x 10^-6 sec

Fig. 14 Yield of transplutonium isotopes from high-flux thermal-neutron irradiation of actinides.

high neutron flux in the resonance energy region, rather than in the thermal region. Computations indicate that fluxes above thermal energy in the range of 10^{14} to 10^{15} n/(cm^2)(sec) might be achieved. Sufficient target material to make effective use of such a large reactor would not be available for some years, when large quantities of ^{243}Am and ^{244}Cm will be separated from spent power-reactor fuels. One estimate indicates that hundreds of grams of ^{252}Cf per year could be produced by a reactor operating in the resonance mode, although neutron-absorption resonance integrals are not sufficiently well known at this time to suggest the accuracy of this prediction.

Underground Nuclear Explosions for Production of Isotopes

Underground nuclear explosives should not be overlooked as a future practical source of these elements since it has the distinct advantage of producing the higher, relatively short-lived isotopes in much greater proportion than can be accomplished by reactor irradiation. Typical reactions are shown in Fig. 15. For example, the largest quantity of the 95-day ^{257}Fm that has yet been produced and recovered was obtained from an underground explosion. A theoretical assessment, made at Los Alamos, of potential yields of heavy nuclides from underground explosions, shows that concentrations of ^{257}Fm of the order of nanograms per ton of debris can be anticipated. Table 2 lists some of the nuclear reactions that could be studied in a laboratory environment if sufficient ^{257}Fm were to be recovered from the site of an underground explosion.

Table 2 Potential Nuclear Reactions of ^{257}Fm

$(n,\gamma)\,^{258}$Fm	$(t,n)\,^{259}$Md
$(d,n)\,^{258}$Md	$(\alpha,n)\,^{260}$No
$(d,2n)\,^{257}$Md	$(\alpha,2n)\,^{259}$No
$(t,p)\,^{259}$Fm	$(n,p)\,^{257}$Es

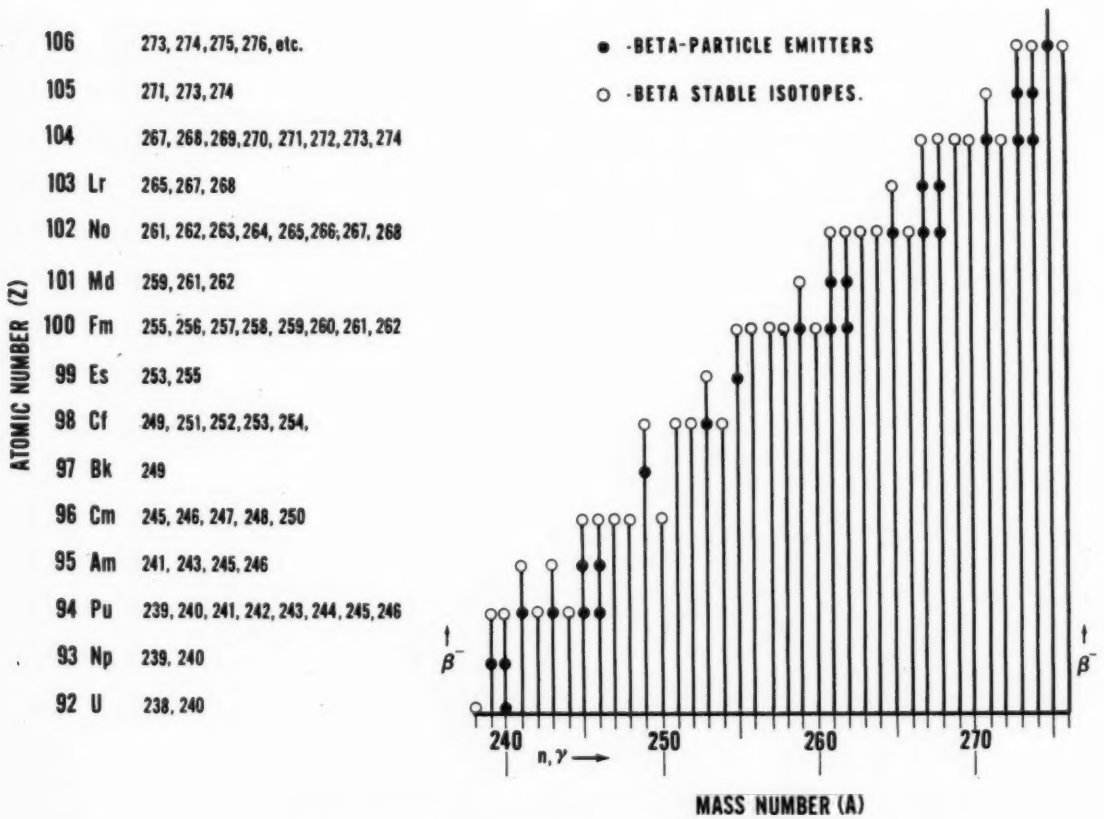

Fig. 15 Isotopes produced in underground nuclear explosions.

The Accelerator as a Neutron Source

Our Canadian colleagues have been speculating on the possibility of still another approach to large-scale heavy-nuclide production. They propose the use of a proton accelerator to produce intense neutron fluxes in a massive target by spallation reactions. Called the Intense Neutron Generator (ING), this very large and expensive machine would produce a proton beam of 65 ma at 1 Gev energy incident on a liquid-metal target. The spallation neutrons would be moderated in a tank of heavy water and the thermalized neutrons would be withdrawn through a beam tube. The design objective of the machine is a thermal flux of 10^{16}. Some of you may recall the efforts of E. O. Lawrence along this line at Livermore almost 20 years ago.

Actinides in the Future

The future for the actinide elements appears to offer continuing excitement, with literally *new worlds* becoming available for their utilization.

Reliable and compact power supplies for manned space missions of perhaps several years' duration could require many kilograms of actinide heat sources in their electric power supplies, with the waste heat being gainfully employed to maintain the quality of the spacecraft's cabin atmosphere and to recycle water for drinking, cooking, and sanitary purposes.

Underwater laboratories—the "Sealabs"—could use similar or even larger power supplies as well as the waste heat generated by them, since providing heat and power several hundred feet below the surface of the ocean for months to years at a time is a true technological challenge.

Many new medical applications for the actinides will appear, I am certain, either in the form of power supplies for use in prosthetic devices of many different sorts or as radiation sources for diagnostic, therapeutic, and research purposes.

The chemists of the years to come will be trained through the educational process to know the chemistry of the actinide elements in the same way that they now master the chemistry of the naturally occurring elements, and they will have available relatively stable isotopes of the actinides with which to perform their experimental research.

A scientific challenge still remaining—although many people have attempted to find a solution with but limited success—is the means for converting the kinetic energy of the emissions of radioisotopes directly into electricity without going through a degradation to thermal energy. Such a process could conceivably have a very high efficiency and would open up new vistas for applications which we probably cannot even visualize now.

I end on this note because it is the duty of someone to sound the clarion for things yet to be done, rather than noting with smugness and satisfaction that everything important has been accomplished already.

(PSB and RHL)

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Section

I

Isotopes and Radiation Technology

Isotope Production and Development

Values in Spent Fuel from Power Reactors*

By C. A. Rohrmann†

Abstract: *The objective of this study was to appraise the magnitude of future amounts of by-products available from discharged reactor fuels, to consider the possible candidate materials that are most likely to have value, to utilize some simplified ground rules for appraising value and recovery cost, and to estimate in a preliminary way the value of candidate materials in unprocessed fuels. The candidate by-products from reactor fuels are considered in three groups: (1) fission products, both radioactive and stable; (2) the higher isotopes, i.e., ^{236}U, ^{237}Np, and isotopes of americium and curium; and (3) the plutonium isotopes. Although the reactor operator may expect only modest credit from fission products, at the level of credit for absorbed neutrons utilized in this study very high returns to the fuel owner or reactor operator are indicated for the other two groups, which could significantly lower the cost of power. This credit appears so large for certain products formed by neutron absorption that a critical analysis of the value of neutrons and the value of products is suggested. Continued laboratory-scale investigation of integrated processes for fuel and isotope processing is recommended, with emphasis on the technology of americium, curium, the noble metals, and the rare gases. Credit for the fissionable plutonium isotopes at the published $10/g figure assures the soundness of the economics of fuel processing.*

Although the recovery of useful fission products and transuranium isotopes from spent reactor fuels has been studied in some detail,‡ the author feels that the unprecedented growth of civilian nuclear power[1] merits a renewed inquiry and reassessment of the worth of many of these potentially exploitable materials. The economic recovery of these isotopes and elements present in discharged reactor fuels should be of interest to a wide spectrum of scientists and engineers, including:

- The reactor designer, who may wish to capitalize on design concepts that favor formation of certain products in highest yields.
- The reactor operator, who may expect credit for neutrons invested in the higher isotopes that are produced unavoidably and whose production can be increased by optimization.
- The fuel processor, who may profit from his efforts to recover these materials.
- The power consumer, who can expect to share in the credits to the reactor operator by obtaining cheaper power.
- The developer of new enterprises through practical application of some of these materials that are available from no other sources, that may be competitive with conventional sources, or that have unique properties to fill new demands.

The results of an earlier study of this subject appear in Ref. 2.

The objectives of the present study are to appraise the magnitude of future amounts of by-products available from discharged reactor fuels, to establish the candidate materials most likely to have value, to utilize some simplified ground rules for appraising value and recovery cost, and to estimate in a preliminary way the value of candidate materials in unprocessed fuels. It is not intended that these estimates be precise but only that they have acceptable and reasonable bases. Such information is considered sufficient to determine if further interest or study is justified. For such a study the author has made the following assumptions:

1. There is an existing market for transuranium isotopes including ^{238}Pu and ^{244}Cm and the other elements and isotopes at the projected prices.

*This paper is a revised (May 1968) version of BNWL-25.

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‡See appended bibliography.

2. A competitive situation exists for reactor fuel processing in combination with isotope recovery systems (more than one fuel processor in business).

3. The products are available as the most appropriate pure solid compounds or aqueous solutions except that the rare gases are marketed as gases and the noble metals as metals.

4. Fission-product radioisotopic heat sources (^{90}Sr, ^{106}Ru, ^{137}Cs, ^{144}Ce, ^{147}Pm) are marketed as bulk products at approximately the HIP (Hanford Isotopes Production Plant Engineering Study[3]) cost for encapsulated sources.

5. Formation and concentration of the products are as established in Refs. 4 to 6 for typical low-enrichment power-reactor fuels.

6. No isotopic separation processes are used for any of the products.

7. Absorbed neutrons are assumed to have a value that is equal to their incremental cost of production. Where credit for neutrons absorbed is used to establish value, neutrons are valued at $2300/g (minimum incremental cost).

8. All fuels will have been irradiated to 25,000 megawatt days per metric ton (Mwd/t).

9. The cost of fuel processing is $27,000/ton of uranium, with one-half of this cost charged to plutonium ($2/g for 25,000 Mwd/t) and the other half to recovered uranium.

10. Both ^{237}Np and ^{242}Cm are considered as precursors of ^{238}Pu and are valued accordingly.

11. Americium-243 is the precursor of ^{244}Cm and is valued accordingly.

12. Other products formed by neutron addition have values determined by the value of the neutrons absorbed, adjusted for the yield of such neutrons in progressing to the ultimate products.

13. The estimate of the cost of producing the materials is based on judgments involving process complexity; degree of process integration with other companion products; value of ultimate product (e.g., the ^{243}Am value is high because it is the precursor of ^{244}Cm, a more valuable product); total value of invested neutrons (see assumption 7); relation between the price of products obtained by conventional industrial chemical processing and the cost of conventional raw materials. In a number of products, this ratio is close to 10.

14. Future prices for products other than ^{238}Pu, ^{244}Cm, and fission-product heat sources are based on judgments of known costs and selling prices and estimates of prices which appear reasonable for the market.

15. The products are materials which are either stable or of sufficiently long half-life and yields to ensure multigram amounts being available per ton of fuel at the time of processing (6 months to 1 year after reactor discharge). In addition, some reasonable expectation for use must be visualized. Unless the product, either an element or an isotope of an element that occurs in nature, is of high value (about $1/g or more), it is not included in the listing of current prices of some costly commercial elements (see Table 5). Materials of low fission yield or low value are thus excluded.

Discussion

Recent forecasts and estimates of the growth of nuclear power are far more optimistic than earlier projections. These estimates indicate the justification for additional fuel-processing plants in the mid-1970's, with a rapidly increasing fuel-processing load in the following 10 years. A nuclear power industry exceeding 150,000 Mw(e) has been forecast by 1980 (Fig. 1). Under such circumstances the amount of many elements and isotopes in fuel-processing wastes may reach such levels and values that, if all could be marketed, the fuel-cycle economics could be appreciably influenced. In addition, there could be a great impact on existing marketing of elements existing in nature, which are also produced in fission, such as rhodium, palladium, and xenon. Rhodium, for example, warrants

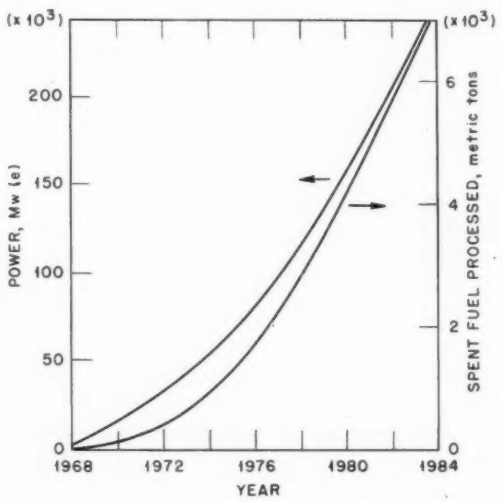

Fig. 1 Estimated growth of nuclear power and spent fuel processing in the United States in the next 15 years (end-of-year basis; Ref. 6, p. 155).

some special comment. Occurring in nature as an element with only a single isotope, it is a member of the platinum group of so-called "noble metals" and is one of the most expensive of all stable solid elements. Yet, even though it is rare and costly, it has such outstanding properties of high-temperature durability and corrosion resistance* that it is a fairly common, important, and useful article of commerce. (For example, a common brand of kitchen gas ranges uses a small coil of Pt–Rh alloy wire as the electric pilot "light" for the gas broiler.†) The most important use of rhodium is, however, the application in which it greatly improves the durability of the platinum gauze catalyst used in ammonia oxidation for nitric acid production. Fully 80% (about 2×10^7 tons) of the world production of nitric acid is used today to produce ammonium nitrate fertilizer. It is readily conceivable that rhodium derived as a by-product of nuclear energy and applied in such conventional ways will make a major contribution to the alleviation of hunger throughout the world. Nuclear energy would thereby not only accommodate man's ever-increasing need for electric power but would also at the same time assist significantly in satisfying man's even more critical need for food.

The short half-lives of most of its artificially produced isotopes ensure that fission-product rhodium will be essentially stable and the same as that found in nature.‡ As a fission product, it falls in the group of lighter isotopes formed in highest fission yield and is only slightly less prevalent than ^{90}Sr. Furthermore, from plutonium fission, it appears in much greater yield than does ^{90}Sr (Fig. 2). Unless some practical recovery technique is found for this valuable element in the fuel-processing wastes, more rhodium will be delivered to underground confinement annually by 1977 than was mined in 1962 worldwide and marketed in the United States! Similarly palladium resulting from fission would also comprise at that time a significant fraction of the palladium mined and marketed today.

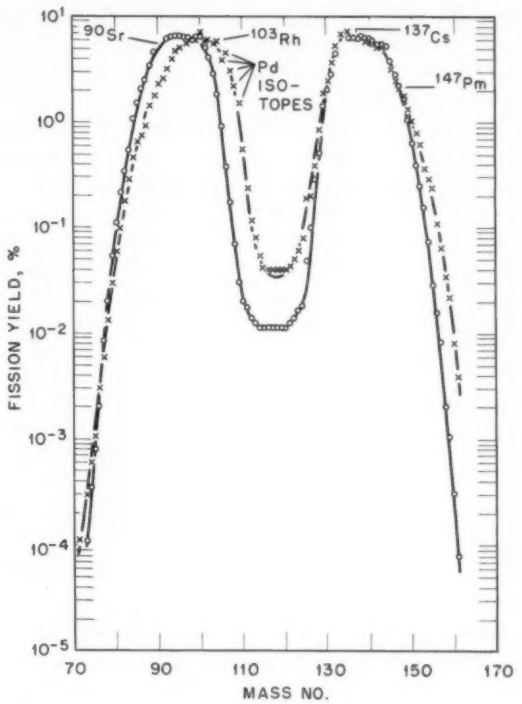

Fig. 2 Yields from thermal fission of ^{235}U (○) and ^{239}Pu (X). [From S. Katcoff, Fission-Product Yields from Neutron-Induced Fission, *Nucleonics*, 18(11): 201 (1960).]

Xenon, now a rare and costly by-product of a few liquid-air manufacturing plants, will also be discharged in large quantities. The annual discharge of xenon from reactor fuel-processing plants by about 1980 will be equivalent to that existing in about 6 cu miles of air. Depending on the fuel-dissolution process used, this xenon could be evolved in fairly high concentrations in off-gases.

These three elements—rhodium, palladium, and xenon—are examples of fission products that are essentially stable elements§ for which expanding markets already exist but which, for rhodium and palladium, most certainly have shrinking world reserves.

*The *Minerals Yearbook* for 1937, p. 752, makes the statement: "Because of its brilliance and durability, rhodium has been called 'The diamond of the metals.' As rhodium is one of the whitest and hardest of all metals and never tarnishes, rhodium plating is being used more and more."

†*Industrial Research*, November 1964, p. 9.

‡An $(n,2n)$ reaction produces active ^{102}Rh of both 206-day and 2.9-year isomers. The concentration of these in power-reactor fuel has not been established, but it is expected to be low enough that important uses can still be considered.

§At least 20% of the palladium should be ^{107}Pd. This is a radioactive isotope of such long half-life and of such low pure-beta energy that it is expected to be used in all normal commercial applications without need for control. Although silver is also formed in fission and at a fairly high yield from plutonium, it is not sufficiently valuable to be seriously considered as a significant by-product. Furthermore, it will very likely be highly contaminated with active isotopes such as 260-day ^{110m}Ag and >5-year ^{108m}Ag.

The situation with the radioactive elements and isotopes is even more exciting (or alarming), since these can be derived only from atomic energy processes. Under the projected conditions, tons of such materials will not only be available but will also have to be safely disposed of. Hopefully, economic, safe, and highly beneficial uses for most of these unique materials will be developed. Among these, there will exist significant amounts of the higher isotopes, which are not produced by fission but by neutron capture in the nonfissioning fraction of the fuels. Among such isotopes is curium. Whereas today a major effort is going on at the Savannah River Plant to produce a 6-kg amount of ^{244}Cm, an approximately equal quantity will have been discharged in about 600 tons of certain conventional power-reactor fuels of high exposure. By 1980, about 60 kg could exist in the output of fuel-processing wastes during that year. Furthermore, it is likely that this will be extractable as a by-product at relatively low cost.

Concurrently the precursors of valuable nuclides such as ^{238}Pu and ^{244}Cm will also be at hand. The availability of these materials will be further enhanced by the recovery and recycle of the precursors, which include ^{236}U, ^{237}Np, ^{240}Pu, ^{241}Pu, ^{241}Am, ^{242}Pu, ^{242}Cm, and ^{243}Am. All of these should have values based on either their utility in producing the ^{238}Pu and ^{244}Cm or at least on the neutron investments involved.

A greatly changed situation will exist when essentially all the plutonium produced in power reactors is recycled. This will involve, especially, a further enhancement of the availability of the higher isotopes. To a lesser extent this is also the case for a few of the potentially valuable fission products that are produced in higher fission yield from plutonium than from ^{235}U, e.g., rhodium and palladium.

This study is concerned essentially with elements or isotopes that will be available annually in kilogram quantities from plants that are processing normal power-reactor fuels of low enrichment. Although many other potentially useful isotopes can also be produced by special target irradiations, the production of such materials is not a part of this review. Special, and possibly useful, isotopes whose fission yield is small and the higher isotopes beyond ^{244}Cm, which would be produced in milligram amounts, are also excluded.

Value of Neutrons

The intriguing prospects of using neutrons to produce unique materials encourage efforts to consider neutrons as a raw material or an essential material in conventional ways for assessing values and manufacturing costs by the usual methods of the chemical processing industry. However, the variety of reactors and the special features of each make the appraisal of neutron cost or value a subject so clouded by the wide differences in bases and situations that, until recently, there has been an apparent reluctance to display any cost figures on the subject. It may have been concluded that neutrons are so expensive that their intentional use for the production of materials other than for defense (plutonium, tritium, etc.) is almost unthinkable. However, some relatively uncomplicated situations can certainly be assumed to provide a basis for placing a dollar value on neutrons. There are also certain isotopes that have such high value when compared with costly, yet conventional, materials that the neutron cost involved in their manufacture could actually be almost insignificant. For example, for certain purposes, a most conventional means of producing a source of neutrons for research purposes is by mixing ^{226}Ra and beryllium to produce a substantial output of neutrons (about 10^7 n/sec per gram of radium present). Over the years radium has been available at about $15,000/g. Any other energetic alpha emitter could also be used in the same way, and several such emitters can be produced by neutron irradiation of certain target materials in reactors. Among these could be ^{228}Th, which has more than 1000 times the alpha activity of ^{226}Ra.

To be competitive with radium for use in these special neutron producers, ^{228}Th would have to be available at a figure not in excess of a seemingly ridiculously high cost of $15 million/g! Technology is actually at hand to ensure product costs far below this level.[7]

Thorium-228 can be produced by irradiation of ^{230}Th. Two grams of neutrons added to 230 g (1 mole) of ^{230}Th produces 1 mole of ^{232}U, which will decay to yield 228 g of ^{228}Th. These neutrons then should be worth 228 x $15 million/2, or about $1.7 billion/g!

A reactor can be visualized in which nearly 1 g of net neutrons (neutrons available for plutonium or other isotope production) could be made available for each gram mole of ^{235}U fissioned. With fully enriched ^{235}U available at about $11.20/g* of contained uranium (~$12/g of 100% ^{235}U), a net gram of neutrons should cost about $2600. This calculation is

*AEC Manual, Appendix 1701—Pricing Handbook, Nov. 18, 1965.

obviously unrealistic since it does not include such costs as fuel and target preparation, reprocessing, and use charges. On the other hand, costly, fully enriched uranium is obviously not the most economical source of neutrons, particularly if the reactor in which the irradiation is taking place is fueled with low-enrichment or natural uranium. Assuming $12/lb for natural-uranium metal (hence containing ^{235}U at $3.75/g), neutrons may cost as little as about $900/g on the same unrealistic basis! However, on such a basis the cost of neutrons needed to produce an isotope with an atomic weight of about 225 need be only about $4/g of the element or isotope. Many isotopes can be mentioned that are valued at much higher figures—for example, ^{242}Cm at $20,000/g (Ref. 8). The conclusion may thus be reached that, in reality, for certain valuable elements or isotopes, the neutron cost component can be minor.

Test or experimental reactors, of course, cannot be considered as the most economical or practical means of producing multigram quantities of any isotope or element, since in such small reactors too large a fraction of the total useful reactor volume and corresponding operating and fixed costs must be charged to the specific isotope-production program involved. With such viewpoints, neutrons in large reactors appear as rather logical "raw materials" for the manufacture of useful products. Similarly any reactor operation that produces recoverable and valuable elements or isotopes intentionally, inadvertently, or incidentally, as by-products could logically be credited for at least the value of the neutrons invested in these materials. Although this view may not apply to fission products, it could be argued to apply to all the higher isotopes that can be shown to have value.

The following assumptions are regarded as reasonable for establishing a likely minimum incremental value for neutrons:

1. A power reactor normally uses fuel of 2% enrichment under conditions of private ownership.

2. An increase of enrichment to 2.02% is needed to provide neutrons for target irradiation to produce special isotopes, with the fuel burned to the same final ^{235}U content.

3. The reactor operates so that a gram mole of fissioned ^{235}U yields 1 net gram of neutrons used for special isotope production.

4. In this operation no extra costs are involved in fabricating the 2.02% fuel or in its final chemical processing.

5. The cost for 2% enriched uranium is $139.15/kg and for 2.02% fuel, $141.12.

For each kilogram of 2.02% fuel used, 0.2 g of ^{235}U is consumed specifically for isotope production. Since 235/0.2 = 1175 kg of 2.02% fuel required for 1 gram mole of ^{235}U, and thus for 1 net gram of neutrons, 1175 ($141.12–$139.15) = 1175 ($1.97) = $2314.

The incremental cost of a gram of neutrons under these conditions is therefore $2314, or rounded off, $2300.

Although such an estimate involves many simplifications, it is realistic since it probably represents the lowest range of neutron value. Only relatively minor changes in value occur at different enrichment levels, e.g., 1 or 3% instead of the 2%. The assumption of 1 g of net neutrons per gram mole of fissioned ^{235}U is also optimistic; lower yields are probably certain with corresponding increase in actual neutron value. However, on the basis of the $2300 figure, minimum credits for higher isotopes are realized. This is regarded as a suitable approach for this study.

By-Product Production and Availability

Although for the predictable future (up to 15 years), nuclear power will undoubtedly be based predominantly on low-enrichment light-water reactors, there will also be modest involvement with plutonium recycle and an emergence of fast reactors late in this period. Depending on the magnitude of the fast reactor development, there would be a significant influence on plutonium recycle, resulting in minimized plutonium recycle in thermal reactors. In addition, in this period of early and rapid growth, few reactors will have reached a steady-state or equilibrium operation. Discharged fuels for processing will be of widely varying enrichments and exposures. These factors complicate any projections of by-product availability.

Although additional basic data would be desirable, those at hand provide a fairly good basis for estimating by-product production and availability. However, the most realistic estimates are not readily derived and, furthermore, may not serve the immediate purposes.

In the course of fuel exposure, a variety of by-product fission products—both elements and isotopes—are produced in a fairly predictable manner. However, these materials cannot be considered to be available for useful application until after the fuel is processed, and even then, for a few products, not until they have been aged to eliminate objectionable contaminants. Thus, in any attempt to translate the magnitude of nuclear power into the magnitude of by-products and their availability, consideration must

be given to fuel composition and residence time in a reactor; aging after discharge and before processing; and retention of certain products for a time, before they can be assumed to be useful. In addition, reactors will be of different types, and fuels discharged from them will be of different compositions and exposure histories.

In this study, simplifications have been made. For example, unless otherwise stated, when a metric ton of discharged fuel is reported to contain 384 g of ^{237}Np, this means that averaged fuel from pressurized- and boiling-water low-enrichment reactors exposed to 25,000 Mwd/t and aged for 1 year up to the time of processing contains 384 g of ^{237}Np. Data for such values are from Refs. 4 to 6. When the availability of a certain by-product is estimated for a specified year, this means that the quantity exists in the waste from all fuels processed in that year. It does not mean that the by-product has been recovered and is at hand but that it is available for recovery from fuel-processing wastes discharged in the specified year. In some cases it also identifies the amounts of useful material recovered, after being held for a time for decay of undesirable contaminants, with or without further processing.

Data from Ref. 6 are plotted in Fig. 3 to show the annual availability of the by-products from fuel-processing wastes. This reference provides data from a critical analysis and estimation of the whole nuclear power and fuel-processing industry in the United States through the year 1990, under a variety of reactor-type combinations and fuel-management concepts. The data are presented for nine combinations, with those from Case 9 of Ref. 6 being generally used in the present study.

The products considered appropriate for a study such as this are the elements and isotopes whose annual output from a reactor fuel-processing plant would be measured in not less than kilogram quantities. These products may be highly radioactive, such as ^{147}Pm; slightly radioactive, such as ^{236}U; or even essentially stable, such as rhodium or xenon. They would include both the fission products and the isotopes produced by neutron absorption in the elements of the fuel. Isotopes that can be produced in significant amounts only by neutron irradiation of special target materials are not included in this study; however, some reference is made to them.

Since many of these products will be useful because of their intense radioactivity, the magnitude of their prospective future outputs is emphasized. From the time the AEC began distributing radioisotopes in 1946, Oak Ridge National Laboratory through November 1963 shipped[9] a cumulative total of about 2.35

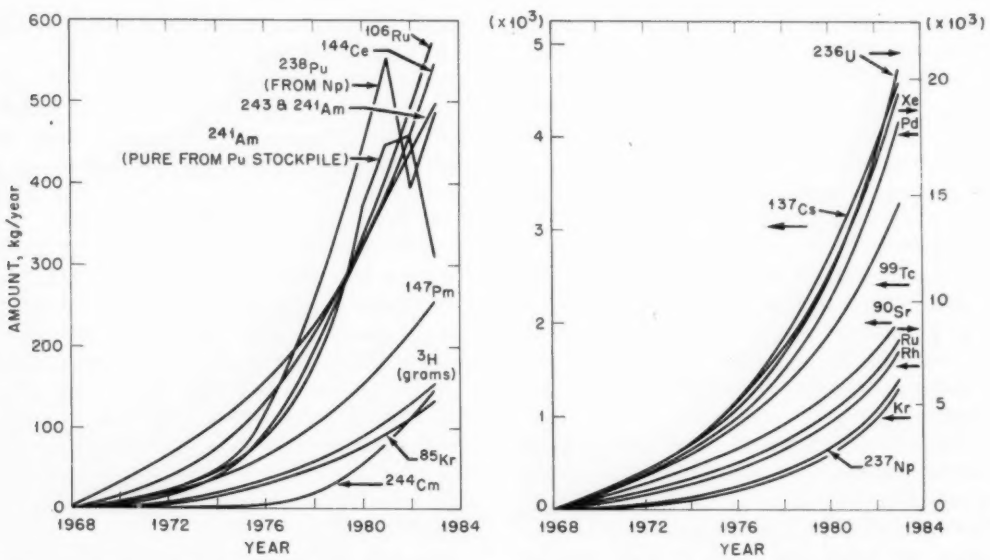

Fig. 3 By-product elements and isotopes from spent nuclear fuel processing in the United States in the next 15 years. (End-of-year basis; Ref. 6, pp. 155-157). [The sharp decline in the curve for ^{239}Pu (from neptunium) and ^{241}Am (pure from plutonium stockpile) exhibits the impact of plutonium diversion to fast reactors around 1980.]

million curies of all isotopes, including 360 kilocuries of ^{90}Sr, one of the commonest radioisotopes produced in highest yield in fission of ^{235}U. In the following 3-year period, the total of these shipments was almost twice that of all previous years. The Hanford Isotopes Plant was proposed[3] to produce 10 million curies of ^{90}Sr/year, along with 140 million curies of three other isotopes. The amount of these same four isotopes alone—^{90}Sr, ^{137}Cs, ^{144}Ce, ^{147}Pm—in reactor fuel discharged[6] in 1980 will be about 4.25 billion curies. These data show the need for the development of uses that can consume these large quantities of radioactive material. Certainly even the well-known and important uses in industry (thickness gages), medicine (radioiodine), and biological research (radiosulfur, phosphorus, etc.) require such small quantities that adequate expansion in these areas cannot be relied upon to consume even an infinitestimal fraction of either present or future outputs. For example, snow gages, which generally use ^{60}Co as the source, contain only 30 to 80 mc per device; and the neutron probes that use ^{226}Ra require only about 5 mc per device. Products that appear to be most reasonable as useful materials are listed in Table 1.

In view of the very large number of elements and isotopes formed by the fission process, the relatively small number selected for consideration in this study calls for further discussion and comment. When one contemplates the typical fission-yield curves (Fig. 2), the very small yield (less than 1%) for a large fraction of all the fission-product elements is impressive. This group includes isotopes of such elements as Zn, As, Se, In, Cd, Ag, Sb, Sn, Gd, Tb, and Dy. It is reasonably clear that expectations for large-scale (kilogram) recovery of such materials are unjustified. This situation, coupled with the practical expectations for producing and, more importantly, finding uses for isotopes of short half-life (even those of high fission yield), also eliminates a very large number of isotopes such as ^{86}Rb, ^{140}Ba, and ^{143}Pr. In addition, many isotopes and elements produced in fission are entirely stable by the time the fuels would be processed and thus are essentially identical with elements plentiful in nature; included among these would be Rb, Y, La, and Pr. It is illogical to consider these as economic products in view of the costly processing necessary for their recovery from the complex mixture of highly radioactive materials present.

If one reviews the available data on fission yields and sums up the yields of the individual isotopes of the elements which are either stable or have long half-lives and then lists all the elements whose total fission yields now exceed 1%, as shown in Table 2, one can readily establish the elements most likely for recovery from power-reactor fuel-processing wastes. A number of the elements from the list in Table 2 have such large yields of radioisotopes that, despite the fact that some of them have fairly short half-lives, they will be highly radioactive for years. These are listed in order of decreasing yield from ^{235}U as:

Zirconium	Krypton
Cesium	Yttrium
Cerium	Tellurium
Ruthenium	Promethium
Strontium	Samarium

Of these, Cs, Ce, Ru, Sr, Kr, and Pm are sufficiently suitable in half-life, yield, and radiation to be considered for practical applications.

After a relatively short aging time (probably within the time that fuel is normally stored and transported to the fuel-processing plant), the following elements, also listed in descending order of yield, would be essentially stable (or exhibit activity of very low penetrating power):

From ^{235}U	From ^{239}Pu
Molybdenum	Molybdenum
Xenon	Xenon
Neodymium	Neodymium
Barium	Palladium
Lanthanum	Barium
Technetium	Technetium
Praseodymium	Lanthanum
Rhodium	Rhodium
Rubidium	Praseodymium
Palladium	Iodine
Iodine	Silver
	Rubidium

Of these, only Xe, Tc, Rh, and Pd are potentially useful, abundant, and valuable enough to be considered for recovery from the wastes of reactor fuel processing.

The considerations indicated above combine to emphasize the probable usefulness and practicability of recovering on a large scale only those fission-product elements and isotopes listed in the first section of Table 1.

The potential availability of heavier isotopes is even more restricted because of the few elements present in fuel which can serve as target materials—simply, uranium and plutonium. It is improbable that elements higher than curium will exist in sufficient concentra-

Table 1 Products from Reactor Fuel Processing That Appear Most Reasonable as Useful Materials

Element or isotope	Half-life, years	Isotopic purity, %	Current price	Potential uses
		A. Fission Products		
^{85}Kr	10.76	5	$22.00/Ci[a]	Special light source, radiation source (phosphors)
^{90}Sr	28	50	0.20/Ci[a]	Heat source, beta source
^{99}Tc	2.1×10^5	100	55.00/g[a]	Corrosion inhibitor, alloying agent, semiconductor
Rhodium	Essentially stable mixture[c]	100	8.00/g[b]	Industrial, electrical, decorative
Ruthenium	Stable mixture +1-year ^{106}Ru	3.3% ^{106}Ru	1.85/g[b]	Heat and beta source
Palladium	Essentially stable mixture[c]	–	1.45/g[b]	Industrial, electrical, decorative
Xenon	Stable mixture	–	35.00/liter (STP)[d]	Special light sources
^{137}Cs	30	35	0.125/Ci[a]	Heat and gamma sources
^{144}Ce	0.78	18[e]	0.15/Ci[a]	Heat and beta sources
^{147}Pm	2.7	100	0.20/Ci[a]	Heat, beta, and X-ray sources
		B. Plutonium Isotopes		
^{238}Pu	87.4	80	$ 1,000/g[f]	Heat and alpha sources
^{239}Pu	24,000	Depends on exposure; can be very high (>95), but most likely 40 to 70	10/g[g]	Fissionable material and source of heavier isotopes
^{240}Pu	6,800	Depends on exposure; 5 to >40	0	Fertile material and source of heavier isotopes
^{241}Pu	13	Depends on exposure; 1 to >20	10/g[g]	Fissionable material and source of heavier isotopes
^{242}Pu	3.8×10^5	Depends on exposure; 0.5 to near 100	0	Target for ^{243}Am formation and source of other heavier isotopes
		C. Heavier Isotopes		
^{236}U	2.4×10^7	A few tenths	–[h]	Target for ^{237}Np formation in normal fuel
^{237}Np	2.1×10^6	Essentially 100	$ 225/g[f]	Target for ^{238}Pu formation
^{241}Am	458	May be 100, but depends on source[i]	1,000/g[f]	Heat source and target for ^{242}Cm (^{238}Pu)
^{243}Am	7,650	May be 100, but depends on source[i]	–	Target for ^{244}Cm and other heavier isotopes
^{243}Cm	0.45	May be 100, but depends on source[i]	20,000/g[j]	Heat source and precursor of ^{238}Pu
^{244}Cm	18.1	Fairly pure; may contain some ^{245}Cm	1,000/g[j]	Heat sources and target for other heavier isotopes

[a] *Radioisotopes, Stable Isotopes, Research Materials,* Catalog available free from Isotope Sales, Building 3037, Isotopes Development Center, Oak Ridge National Laboratory, Post Office Box X, Oak Ridge, Tenn. 37830.

[b] Current prices (calculated from prices per troy ounce), *E/MJ Engineering and Mining Journal,* 169(2): 24 (February 1968).

[c] Palladium includes very-long-lived, very-low-beta-energy ^{107}Pd, assumed to be completely nonhazardous and nonobjectionable in all uses. Rhodium will include slight but significant amounts of active ^{102}Rh. The concentrations in power-reactor fuels have not yet been established.

[d] Price of by-product material from liquid-air manufacturing.

[e] For low exposures; for long exposures (25,000 Mwd/t) and a 1-year decay time before processing, the composition is about 4.5%.

[f] *Nucleonics Week,* 9(17): 4 (Apr. 25, 1968).

[g] The estimated fuel value of ^{239}Pu and ^{241}Pu is $10/g (Ref. 10).

[h] Not separated; stays with ^{238}U or enriched uranium.

[i] Pure ^{241}Am may be recovered from aged plutonium (from ^{241}Pu decay); however, from reactor fuel processing it will be mixed with ^{243}Am and even some ^{242}Am (152 years); the composition of such mixtures depends on the exposure. At 25,000 Mwd/t exposure, the americium is essentially 50-50 ^{241}Am and ^{243}Am. Such mixtures also assure that separated curium will also consist of both ^{242}Cm and ^{244}Cm, but on aging fairly pure ^{244}Cm should result along with pure ^{238}Pu from the decayed ^{242}Cm.

[j] H. L. Davis, Radionuclide Power for Space—Part 1, Isotope Cost and Availability, *Nucleonics,* 21(3): 61-65, especially p. 65 (March 1963).

Table 2 Elements with Fission Yields Greater Than ~1%

From ^{235}U		From ^{239}Pu	
Element	Yield, at.%	Element	Yield, at.%
Zirconium	31.4	Molybdenum	23.6
Molybdenum	25.30	Xenon	23.2
Xenon	21.80	Ruthenium	22.2
Neodymium	19.98	Cesium	20.8
Cesium	19.2	Zirconium	19.15
Cerium	12.10	Neodymium	17.8
Ruthenium	11.28	Palladium	16.75
Strontium	9.30	Cerium	10.60
Barium	6.65	Barium	6.30
Lanthanum	6.40	Technetium	6.10
Technetium	6.25	Lanthanum	6.0
Praseodymium	5.75	Rhodium	5.60
Krypton	4.85	Praseodymium	5.60
Yttrium	4.70	Strontium	3.60
Rhodium	2.90	Tellurium	3.42
Tellurium	2.60	Samarium	2.97
Rubidium	2.50	Promethium	2.05
Promethium	2.15	Krypton	2.05
Samarium	1.75	Iodine	1.77
Palladium	1.52	Yttrium	1.70
Iodine	1.25	Silver	1.55
		Rubidium	0.91

tion to justify their recovery from normal fuels. Feasible but costly methods, e.g., further irradiation of a target material such as recovered curium, would, however, be a more practical means of producing those elements beyond curium.[11] It is conceivable that power-reactor operators may even find circumstances under which they could profitably charge for such irradiation services. Such possibilities are, however, beyond the scope of this present study. Table 1 also lists the heavier isotopes which are certain to be of interest and which will be present in appreciable amounts, as well as the plutonium isotopes that would also be valuable precursors of costly americium and curium.

For the noble metals, essentially current prices were assumed for the future.

Product Costs and Pricing

Although several of the products listed in Tables 1 and 2 have been recovered on a fairly large scale from fuel-processing wastes, the concept of an integrated facility to recover each of them routinely and economically has not received intensive study. The problem of establishing values, costs, and prices is therefore one of guessing, or preferably one of estimating on the basis of reasoning and judgment, using whatever information most closely approximates the situation at hand.

No production cost was assumed for ^{236}U since it accompanies massive amounts of ^{238}U and would not be separated. The plutonium isotopes were assumed to bear an equal share of the rather modest processing costs, which were apportioned on a dollars-per-gram basis.

Heat-Source Fission Products

The processing of ^{90}Sr, ^{137}Cs, ^{144}Ce, and ^{147}Pm, as presented in the study of the proposed Hanford Isotopes Plant,[3] gave a fairly good basis for costs from which other fission-product production costs could be estimated, based on throughput (grams per ton of fuel) and anticipated processing complexity. The costs of the products from this plant are, however, assumed to be for bulk products, unencapsulated. The assumed production cost, including profit, for many of these would be about 90% of the future bulk price, with 10% of this figure being the amount creditable to the value of the unprocessed fuel. The 10% figure can be supported by a review of several common industrial chemical processing situations; for example, market price*-to-raw concentrate costs show the following ratios:

Conversion	Price-to-cost ratio
Barite to precipitate $BaCO_3$	7.85
Beryllium ore to BeO	8
Chromite to CrO_3	10.75
Fluorspar to HF (aq.)	9.6
Phosphate rock to ammonium phosphate (food grade)	9.8
Monazite to rare-earth oxides (CeO_2)	10
Zircon to ZrO_2 (optical grade)	9.9

For those intermediates or precursors in which the only investment is the value of neutrons absorbed, such neutrons are charged in at $2300/g, which is regarded as a low incremental cost of a typical gram of neutrons. This procedure was modified for ^{240}Pu, since here a portion of the invested neutrons goes to produce an isotope that is largely consumed in fission (^{241}Pu) instead of progressing up the scale to ^{242}Pu.

Where the use of these four fission-product nuclides (^{90}Sr, ^{137}Cs, ^{144}Ce, and ^{147}Pm) relates to their application as heat or power sources, the future

*From current market quotations in *Oil, Paint, and Drug Reporter*, and *E/MJ Engineering and Mining Journal*.

the total costs, including feed, as reported for *encapsulated* products in that reference. Although no feed cost is expected,* it is retained and assumed to cover amortization, taxes, and profit. The assumed future production cost is then estimated to be about 90% of the future prices, as mentioned above.

Power-reactor fuels will tend toward longer exposures. Furthermore, there probably will be no great incentive to ensure a short cooling time between reactor discharge and chemical processing. These two factors introduce variables that greatly influence the quality of certain by-product elements and isotopes. In some cases higher yields will result from prolonged aging; in others, lower yields and lower concentrations will be obtained. Cerium-144 is a good example of the latter situation. Its rather short half-life leads to saturation concentrations at moderate exposures. In addition, the concentration of the stable isotopes of cerium continues to increase throughout the exposure period, leading to lower and lower isotopic concentrations of ^{144}Ce. Finally, at the end of the exposure period and during the aging, cooling, or storage period between discharge and chemical processing, the ^{144}Ce decreases through beta decay, leading to a further major reduction in isotopic concentration. Thus, to achieve highest ^{144}Ce yields and isotopic concentrations, the fuel exposure should not exceed the minimum level at which saturation is achieved, and the aging period should be as short as possible. Neither of these conditions is compatible with power-reactor fuel-cycle economics. Although ^{144}Ce is an outstanding example of this problem, ^{106}Ru presents very much the same situation. For ^{106}Ru, however, an advantage exists because of the much higher ^{106}Ru fission yields obtainable from operations that emphasize the burning of plutonium. Also, the longer half-life slightly favors the buildup of ^{106}Ru. However, the buildup of inert ruthenium isotopes is also very significant. From data in Ref. 4, at 25,000 Mwd/t for low-enrichment ^{235}U fuels, it can be shown that 1 year after reactor discharge the cerium will contain about 4.5% ^{144}Ce, and the ruthenium, about 3.3% ^{106}Ru.

Other Fission Products

Krypton-85. For isotopes like ^{85}Kr the establishment of a basis for a future price is very uncertain. At one time ^{85}Kr was expected to be marketable at $15/curie. The current price is $22/curie. The intended uses that supported such expectations have obviously not developed, even with a price of $7.50/curie, which prevailed for 2.5 years before the current price was established. The future price, for the purposes of this study, was assumed to be $1/curie (or about $450/g), which is only about 5% of the current price. It is thought that such a major drop in price may encourage sufficient uses under future conditions and also allow for about an equal sharing of production costs between ^{85}Kr and xenon.

Although, on an annual basis, kilogram quantities of ^{85}Kr should be available, the amount in each ton of fuel is the smallest of any of the fission products of interest. The unique application (phosphors) that originally was expected to support a high price may still be contemplated and may be significantly encouraged by a much lower price. Even at prices one-twentieth of that of current prices, and with rather substantial processing costs, a sufficient margin remains to ensure a significant value for unprocessed fuel. The total krypton in fuel will be about 257 g/ton for 25,000 Mwd/t ^{235}U fuel and about half this value for ^{239}Pu fuels.

Xenon. In view of the very high prices for xenon today, a much lower price was assumed for the future. The very large availability of the fission-product source and the potentially convenient and rich concentrations in the off-gases from fuel processing, coupled with concurrent production of krypton, support expectations for relatively low production costs.

Xenon will be discharged as a mixture of the stable isotopes. The potentially contaminating radioactive xenon isotopes have such short half-lives that by the time fuel is processed they should be essentially absent. The availability of xenon in grams per ton of fuel is far higher than that for any other potentially useful fission products—3987 g/ton of 25,000 Mwd/t fuel. This, plus the feasibility of a fuel-treatment process that could release xenon along with krypton in fairly high concentrations, encourages the consideration of a recovery cost and selling price substantially below that existing today for the small-volume liquid-air by-product. The production of xenon is essentially the same for either ^{235}U or ^{239}Pu fission.

Technetium-99. Technetium, an element not found in nature, is produced by fission in very high yield—50% more than ^{90}Sr from low-enrichment power-reactor fuels. Its isotopes are generally of very short or very long half-lives, the long-lived ones being

*It is even conceivable that fission-product recovery might receive credit for waste-management expenses that would be avoided through sale of these materials.

prices for bulk products are assumed to be essentially adjacent. The principal isotope produced in fission is ^{99}Tc with a half-life of 2.1×10^5 years. The ^{98}Tc, which has an even longer half-life (1.5×10^6 years), is not produced in yield high enough to be of interest. The other fission-produced technetium isotopes have half-lives measured in minutes or less. Technetium-99 would therefore be obtained in high isotopic purity. Its recovery by either solvent extraction or ion exchange is simple and has been demonstrated on a kilogram scale. The ion-exchange process used and the yields are so similar to those for ^{137}Cs that the costs per gram have been assumed to be about the same.

Uses for technetium have not been developed. Its existence as a new element, the trend toward lower costs, and some of its properties—particularly that of providing corrosion resistance in steel exposed to aqueous solutions containing small amounts of technetium—suggest future uses. Its use as an alloying agent to improve the metallurgical properties of tungsten and its superconductivity characteristics are also being studied. Its long half-life and low-energy pure-beta emissions (free of gamma activity) indicate that it may be used with a minimum of control.

Noble Metals. The similar noble metals—ruthenium, rhodium, and palladium—have the next highest availability (after xenon) of all the potentially useful fission products and might be easily recovered as a group. This possibility would appear to be greatly enhanced in an integrated fuel-processing and multiple-isotope-recovery facility. The cost per gram might then be sufficiently low to make recovery economically competitive with conventional ore sources. The recovery cost is therefore assumed to be only slightly below the expected selling prices.

In precious-metal mining today, the existence of accessible and large hard-rock ore bodies containing about ½ oz gold/ton ($17) justifies mining and processing. For large placer deposits, gold values at far less than 1 g/ton ($1) justify mining. In expected power-reactor fuels, the rhodium value alone based on the present prices will be about $2500/ton. Palladium values would add about another $1000/ton of fuel. Compared with the concentrations in natural ores, such levels of the noble metals in spent fuels would be regarded by miners and economic geologists as nothing less than fantastic. The acknowledged existence of such concentrations in extensive and available geological formations would certainly initiate a "gold rush" of wild intensity! However, the reality of the high cost of processing intensely radioactive materials, plus the rather modest quantities of such material which can be anticipated in the near future (within the next 5 to 10 years), necessitates a temperate and more deliberate approach to recovery. Although such recovery has not been demonstrated on a significant scale even experimentally, the possibilities and eventual practicality of a large-scale approach seem reasonable as well as probable.

Rhodium-103. Stable fission-product ^{103}Rh is the daughter of 40-day ^{103}Ru. With normal ^{235}U or ^{239}Pu fuel, its yield is about 2.9 or 5.8%, respectively (Fig. 2). The fairly long-lived (3-year) ^{102}Rh may be present in very small amounts as a result of $(n,2n)$ reactions with ^{103}Rh. Very-long-exposed fuels may thus have sufficient ^{102}Rh to impair the unrestricted use of the stable ^{103}Rh. In contrast to this possibility, the cross section for this particular $(n,2n)$ reaction may be so low or the absorption cross section for the resulting ^{102}Rh so high that no problem with ^{102}Rh will arise. Until this question is resolved by precise analyses of samples of fuels exposed for long burnup times, no problem is assumed at this time in the use of rhodium recovered as a fission product.* For fuels in which ^{239}Pu supplies a large fraction of the fissions, the yield of rhodium will be highest. For conventional fuels at 25,000 Mwd/t exposures, the yield is about 337 g/ton (about 80% of the yield of ^{90}Sr). Rhodium from conventional mineral sources today sells for about $8/g.† In the period 1955 to 1964, the average annual sales of rhodium in the United States amounted to about 26,600 troy oz (827 kg). In the rapidly expanding nuclear power industry, rhodium will therefore become increasingly significant as a potentially recoverable and valued by-product.

Little is known of the exact state and fate of rhodium throughout the nuclear fuel separation processes, but a modest analytical effort is being made to obtain such information. The technology of recovering gross quantities of rhodium economically in high yield and purity from conventional fuel-processing wastes has also yet to be developed; however, studies are under way. Some special processes have been patented for recovery of rhodium.[12] One patent relates to the

*These analyses are in process at the present time, but results are not yet available.

†It is of interest to note that this value comes close to that for plutonium from high-exposure power-reactor fuels (with the plutonium valued only at its fuel value of $10/g for ^{239}Pu and ^{241}Pu). Such plutonium can be 60% ^{239}Pu plus 10% ^{241}Pu.

separation of ^{103}Ru, which is aged and then processed for recovery of pure ^{103}Rh; another relates to conversion of nitric acid waste solutions to chloride solutions, from which the rhodium is recovered by sulfide precipitation.

The principal uses of rhodium are in the chemical, glass, electrical, and jewelry (and decorative) industries. In addition to its greatest single use, i.e., improving the durability of platinum catalysts for ammonia oxidation to nitric acid, its alloys with platinum have distinctive properties that are useful in applications involving very high temperatures in air: thermocouples, heating elements, crucibles, etc.

Palladium. Palladium is the most abundant of the noble metals—about 20 times that of rhodium but less than twice that of platinum. Its selling price today exceeds $1.45/g and has been increasing significantly over the last few years. Palladium has widespread use in chemical (catalyst), electrical, glass, dental, and jewelry (decorative) applications. Its use as an electrical contact material in telephone switchgear (relays) is the most important single application. In fission the combined yield of the four principal isotopes is over twice that of either rhodium or ^{90}Sr. As with rhodium, the highest yields are obtained in plutonium fission. At 25,000 Mwd/ton, normal ^{235}U fuels are expected to yield about 875 g of palladium per ton. From plutonium fuels the yield will be about 1300 g/ton. Its price is, however, borderline as a single element of significant value in spent nuclear fuels. With low recovery costs (a possibility with an integrated recovery process for all the noble metals), its recovery would probably be economic. Among its isotopes, there appear to be none that could interfere with uncontrolled use. Radioactive ^{107}Pd, which is about 20% of the isotope mixture, has a long half-life (7 million years) with no gamma emissions and with a beta emission of such low energy (0.035 Mev) that it may be regarded as without hazard for any of the major uses, even dental.

As in the case of rhodium, little is known about palladium in the fuel-recovery process, but analysis of typical high-exposure power-reactor fuel samples is in progress to establish the concentration of palladium and its isotopic composition.[13] Similarly the technology needs development before economic recovery can be ensured. The future magnitude of the nuclear fuel-processing industry could provide amounts of palladium to satisfy a significant fraction of today's growing needs.

Ruthenium. Unfortunately ruthenium has an active isotope, ^{106}Ru, of fairly long half-life (1 year). The realization of conventional uses for such ruthenium is therefore questionable. Ruthenium is one of the most abundant solid elements of potential value in spent fuel. Ruthenium from conventional mineral sources today sells for about $1.85/g. It is assumed to be readily recoverable from spent fuel and to eventually have some uses. If it could be recovered at the indicated costs, it might have potential as a powerful (high power density) radioisotopic heat or power source and thus may be worth possibly 25 to 50 times as much as it would be as a noble metal. The extremely energetic beta-decay energies of its daughter, ^{106}Rh, also suggest potential applications. The fission yield of ^{106}Ru is, however, quite low from ^{235}U (0.38) but from ^{239}Pu fission is a factor of >13 times as great.

Xenon and the noble metals rank among the most expensive of the conventional elements. However, their prices are low compared with the possible costs of most processed radioactive materials. Thus, to be economic for recovery, the processing cost must also be low. Since on a weight basis xenon and the noble metals comprise by far the greatest of the potentially useful products, fairly low recovery costs have been assumed; however, Table 3 shows that the processing cost is only slightly below the assumed future value of these materials. The credit that can be expected is thus also modest.

Heavier Isotopes

For the heavier isotopes that are one step away from the valuable ultimate products (^{236}U for ^{237}Np, ^{243}Am for ^{244}Cm, and ^{241}Am for ^{242}Cm and ^{238}Pu), the value of the ultimate products and a reasonable processing cost were taken into consideration in arriving at the value of the intermediates. The value of invested neutrons is also a major factor in assessing the value of these products. Except for ^{237}Np, the heavier isotopes, formed by neutron absorption in the basic fuel materials, uranium and plutonium, are mixtures of isotopes not considered to be separable and include ^{236}U; two americium isotopes, ^{241}Am and ^{243}Am; two curium isotopes, ^{242}Cm and ^{244}Cm; and five plutonium isotopes, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu. For the purpose of this review, the plutonium isotopes will be discussed in detail as a group. Because of the special situation involving the competing neutron reactions of absorption and fission, especially for ^{241}Pu, particular consideration should be given to individual discussions.

Uranium-236. Uranium-236 is formed through neutron absorption by ^{235}U. About 15% (one-sixth) of the neutrons captured by ^{235}U form ^{236}U; the balance result in fission. The ^{236}U thus formed remains with the processed uranium. As with essentially all such neutron reactions, the actual cross sections depend greatly on neutron energy and reactor conditions; the 15% absorption figure is for illustration only.

Uranium-236 is a precursor of ^{237}Np, which is the target material used in the production of valuable and costly ^{238}Pu. Uranium-236 is therefore valuable in this production chain. Since it is not fissionable by thermal neutrons, essentially all of it is useful for ^{237}Np formation. Its content in recovered uranium is so low (about 0.2%) that it is assumed not to share in the uranium processing cost.

Realistically its share of any processing cost, figured on a gram basis, would be so small (1.5 cents) compared with its value that it can be neglected. However, since one neutron is invested in its formation, it is assumed to carry an appropriate value, this being the incremental cost of this neutron. With incremental neutrons valued at $2300/g and each gram of neutrons thus distributed over 236 g of ^{236}U, the value of the ^{236}U in the unprocessed fuels is set at $9.75/g ($2300 ÷ 236 = $9.75). This value, rounded to $10, is assumed then to be creditable to the reactor operator for the ^{236}U which he has inadvertently produced.

Neptunium-237. Neptunium-237 originates from two sources: neutron absorption in ^{236}U and the $(n,2n)$ reaction with ^{238}U. For power-reactor fuels of higher exposure, the contribution from the $(n,2n)$ reaction is the smaller. Since ^{237}Np is useful primarily as the target material for the production of valuable ^{238}Pu, its value should reflect not only the investment in neutrons required but also the value of the product ^{238}Pu, the conversion efficiency, and the cost of the chemical processing required to recover the ^{237}Np. The present price of ^{237}Np in gram quantities is $225/g (Table 1) from the USAEC.

Neptunium is an alpha emitter and would be most conveniently handled as such, with a minimum of shielding. The purification is therefore important. In view of the relatively small quantities recoverable from fuels, it would be expected to be more costly than the gamma emitters or the more plentiful alpha emitters such as plutonium, which share its costs with the uranium fuel-processing costs.

Based on the projected price for ^{238}Pu of $250/g, the rather poor yield in going from ^{237}Np to ^{238}Pu (60 to 80%), and the special target preparation, irradiation, and processing for ^{238}Pu and recovery of unconverted ^{237}Np, a figure of $75/g was considered suitable for the future price of ^{237}Np. In January 1966 the British (UKAEA, Amersham) released a price schedule that offered ^{237}Np at $75/g minimum when purchased in amounts of 20 kg. Allowing a rather high figure of $55/g as the production cost, its value in the unprocessed fuel is $20/g; this is essentially equal to the value of the neutrons required for its formation.

Americium. The two principal americium isotopes, ^{241}Am and ^{243}Am, are derived exclusively from successive neutron absorption in ^{239}Pu. Americium-241 is subsequently formed by beta decay of 13-year ^{241}Pu. Americium-243 comes from neutron absorption in ^{242}Pu. Both americium isotopes are long-lived alpha emitters. Americium-241 has a sufficiently short half-life (458 years) that it could be used as a heat source if pure enough and cheap enough. High purity could be obtained by simple reprocessing of pure, aged plutonium. But, unless a low inventory charge prevailed, low-cost americium produced in this way could not be ensured. The isotopic composition of americium obtained from direct processing of power-reactor fuel depends on age and exposure of the fuel. At about 25,000 Mwd/t the isotopic ratio is essentially 50% ^{241}Am and 50% ^{243}Am for low-enrichment ^{235}U fuels. It is noteworthy that at the time of processing fuels of this exposure the total yield of americium is about 175 g, which is substantially greater than that for promethium and other familiar fission products—^{106}Ru, ^{144}Ce, or ^{85}Kr (Table 3).

Irradiation of a target mixture of americium isotopes recovered from power-reactor fuels would produce both ^{242}Cm and ^{244}Cm. After a few years' aging of this mixture of curium isotopes, essentially all the ^{242}Cm would be converted to ^{238}Pu. Thus ^{241}Am, like ^{237}Np, can be considered a source of ^{238}Pu. There may be definite advantages in yield and purity of the ^{238}Pu produced by such a route. Americium-243, on the other hand, is useful only as the immediate precursor of valuable and costly ^{244}Cm; but, in such a process, small quantities of plutonium high in ^{244}Pu could also conceivably be obtained.

For use as a competitive heat source, ^{241}Am must have a value of about one-fifth of that of ^{238}Pu, or $50/g. Its recovery as a by-product of operation of the chromatographic ion-exchange system for promethium production should provide product of required purity

Table 3 Estimated Values of Some Elements and Isotopes from Spent Fuels from Power Reactors

Element or isotope	Potential use	Half-life, years	Specific power, watts/g	Current prices			Assumed future price, $/g[a]	Assumed production cost, $/g	Estimated value in fuel, $/g	Concentration in fuel, g/ton at 25,000 Mwd/ton[b]	Estimated value, $/ton spent fuel	
				$	Unit	Calculated to $/g						
^{85}Kr	Special light and radiation source	10.4	0.54	22.00	Curie[c]	8,930	450	425	25	16 (257 g for total Kr)	400	Fission products $6,786
^{90}Sr	Heat and beta source	28	0.96	0.20	Curie[c]	28.40	20	18	2	400	800	
^{99}Tc	Corrosion inhibitor, alloying agent	2.1×10^5	–	55.00	Gram[c]	55	9	8	1	628	628	
Rh	Industrial, electrical, decorative	Stable	–	8.00	Gram	8.00	8	6	2	337	674	
Ru[f]	Heat and beta source	Stable (+1-year ^{106}Ru, 3.3%)	33.1	1.85	Gram	1.85	4 (as a heat source)	3.50	0.50	1,707 (59 g for ^{106}Ru)	854	
Pd[f]	Industrial, electrical, decorative	Stable	–	1.45	Gram	1.45	1.50	1.00	0.50	876	438	
Xe[f]	Special light source	Stable	–	35.00	Liter[g]	6	2	1.70	0.30	3,987	1,196	
^{137}Cs	Radiation source	30	0.42	0.125	Curie[c]	11	8	7	1	928	928	
^{144}Ce	Heat and beta source	0.78	25.6	0.15	Curie[c]	477	17	15	2	99[h]	198	
^{147}Pm	Heat, radiation, and X-ray source	2.6	0.33	0.20	Curie[c]	186	75	65	10	67[i]	670	
^{238}U	Fertile material (plus residual ^{235}U)	4.5×10^9	–	–	–	–	6.75/lb	6.75/lb	0	9.7×10^5	0	The higher isotopes $44,578
^{236}U	Target for ^{237}Np	2.4×10^7	–	–	–	–	10	0	10[j]	3,169	31,690	
^{237}Np	Target for ^{238}Pu	2.2×10^6	–	225.00	Gram[e]	225	75	55	20[j]	384	7,680	
^{241}Am	Heat, alpha, and X-ray source (also target for ^{242}Cm)	458	0.1	1,000.00	Gram[e]	1,000	115	85	30[j]	56	1,680	
^{242}Cm	Heat source (decays to ^{238}Pu)	163 days	120	167	Watt	20,000[d]	150	110	40[j]	13	(520)[j]	
^{243}Am	Target for ^{244}Cm	7,650	–	–	–	–	130	80	50[j]	57[i]	2,850	
^{244}Cm	Heat and alpha source	18.1	2.65	–	–	1,000[d]	170	110	60[j]	11.3[i]	678	

^{238}Pu	Heat and alpha source	87.4	0.56	1,000.00	Gram	880[e]	250	210	40[j]	(From ^{242}Cm above)	520	
^{239}Pu	Fissionable	2.4×10^4	–	10	Gram	10	11	2	9[j]	5,300	47,700	Plutonium isotopes $84,775
^{240}Pu	Fertile (and source of ^{241}Pu)	6.8×10^3	–	–	–	–	6.50[k]	2	4.50[j]	2,110	9,495	
^{241}Pu	Fissionable (and source of ^{241}Am)	13	–	10	Gram	10	18[k]	2	16[j]	1,005	16,080	
^{242}Pu	Target of ^{243}Am	3.8×10^5	–	–	–	–	38[k]	2	36[j]	305	10,980	
										Value: per metric ton	$136,159	
										per pound	$61.80	
										per kilogram	$136.16	
										per kilowatt-hour (electrical)[m]	0.68 mill	

a. 100% basis, isotope or element in bulk form.

b. These numbers are approximate and vary with reactor spectrum; for plutonium and higher isotopes, they are averages from BWR and PWR without plutonium recycle; per metric ton from Refs. 4, 5, and 6.

c. Radioisotopes, Stable Isotopes, Research Materials, Isotope Sales, Building 3037, Isotopes Development Center, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, Tenn. 37830.

d. Nucleonics, March 1963, p. 63 (Ref. 8).

e. Nucleonics Week, 9(17): 4 (Apr. 25, 1968).

f. Would be mixed isotopes, essentially stable (except ruthenium); some have high fission yields, especially from ^{239}Pu.

g. Price for by-product material from liquid-air manufacturing.

h. One year after discharge.

i. One half-life after discharge.

j. These values are based on credit for absorbed neutrons (at $2300/g neutrons effective in the production of higher isotopes).

k. These assumed future prices are substantially greater than those now generally accepted as the fuel value of plutonium isotopes.

l. As ^{238}Pu, value included with plutonium isotopes.

m. At $33^1/_3$% efficiency thermal to electrical.

at low cost but probably not as low as $50/g. The direct accumulation in the promethium system from which it must be removed thus obviously indicates recovery with little effort. If it is to share the cost of operating the ion-exchange system, it should not be cheaper than the per-gram cost of promethium. Therefore $85/g seems reasonable for the production cost of ^{241}Am. Since ^{243}Am would be recovered concurrently with ^{241}Am, its production cost is assumed to be about the same, or $80/g. With the value of invested neutrons at $30 and $50, respectively, the assumed future prices would be $115/g for ^{241}Am and $130/g for ^{243}Am.

Curium. The two principal curium isotopes found in spent fuel are ^{242}Cm and ^{244}Cm. Because of the relatively short half-life of ^{242}Cm (163 days), the aging time before processing is important in establishing the ratio of these two isotopes. Both are derived from neutron capture by the two americium isotopes. As mentioned above, ^{242}Cm could be recovered as a step in the production of ^{238}Pu.

For the ^{244}Cm to be useful as a heat source, it must be aged sufficiently long to ensure a low enough ^{242}Cm content so that the heat output would be predominantly from long-lived ^{244}Cm. Otherwise the heat source would show the rapid decline in heat output characteristic of ^{242}Cm. The aging time required would be dependent on the concentration of the ^{242}Cm; however, for anticipated concentrations, 2 to 4 years would be required to lower the ^{242}Cm content by decay to less than 0.25% (at this level, less than about 10% of the total initial heat of the source would be coming from the ^{242}Cm).

Such an aging step would, of course, involve some loss of ^{244}Cm (probably less than 15%). However, such losses are much smaller than those resulting from the required aging of heat sources such as promethium. The concentrations of ^{244}Cm and aged by-product promethium, when compared on a weight basis, show the curium concentration to be about one-sixth of that of promethium. However, the significance of the ^{244}Cm concentration—about 11 g/ton from low-enrichment ^{235}U fuels (e.g., average for pressurized-water and boiling-water reactors)—is more impressive when the comparison is made on the basis of the emitted energy: about 2.65 watts/g for ^{244}Cm vs. 0.33 watt/g for ^{147}Pm. This ^{147}Pm concentration assumes aging for about one half-life to remove the offensive (gamma-emitting) ^{148}Pm. Therefore, effectively, only one-half of the concentration of ^{147}Pm in the fuel as discharged can be considered useful as a heat source. This assumption assures about 67 g of promethium per ton. This situation, coupled with the inherent low specific power of promethium, gives a different and more favorable picture of the comparative significance of the ^{244}Cm concentration. From this standpoint, then, ^{244}Cm will yield more useful heat than ^{147}Pm:

^{244}Cm = 11.3 g/ton of fuel; at 90% recovery and 2.65 watts/g = 26.9 watts/ton of fuel

^{147}Pm = 67 g/ton of fuel; at 90% recovery and 0.33 watt/g = 19.9 watts/ton of fuel

A similar comparison for ^{238}Pu based on its production from the neptunium recovered from the same fuel (Table 3) shows:

For 384 g ^{237}Np/ton of fuel, 90% recovery, and 65% conversion efficiency to ^{238}Pu (in the form of mixed plutonium isotopes containing 80% ^{238}Pu) having a specific power of 0.56 watt/g of ^{238}Pu:

(384)(0.65)(0.9)(0.56) = 126 watts/ton of fuel

Thus the heat available from ^{238}Pu, which was produced by an even more circuitous and time-consuming route, is nearly 5 times that obtainable directly from by-product ^{244}Cm. However, this route for ^{238}Pu takes time since the neptunium must be irradiated in cycles, each of which converts only a fraction of the neptunium to ^{238}Pu; then the mixture must be separated and purified and the neptunium refabricated into target elements for the next irradiation cycle. Furthermore, under conditions of plutonium recycle the average production of ^{244}Cm is nearly 20 times that shown in Table 3.

Although under anticipated practical conditions little ^{242}Cm is expected to exist in fuels at the time of processing (1 year after discharge), whatever quantity does exist at the time will be recoverable as ^{238}Pu from the aged ^{244}Cm.

The future price or value of ^{242}Cm has therefore been placed at $150/g. This is far below projections referenced in Table 1 and has been set at this level because of both the anticipated low cost of the target, ^{241}Am, and its possible use to produce ^{238}Pu. Therefore ^{242}Cm would have to be valued at appreciably less than ^{238}Pu. Although curium, like americium, can be recovered from the ion-exchange system, the smaller quantities involved and the probable requirement for special procedures to cope with the intense and damaging alpha activity justify a substantially higher production cost for the ^{242}Cm, but the credit for ^{242}Cm in fuel should also be based on its use as a source of ^{238}Pu. A production cost of

$110/g was thus concluded as reasonable for ^{242}Cm. Fairly pure ^{242}Cm for heat-source uses must, however, be specially produced by irradiation of fairly pure ^{241}Am and at a much higher cost.

The future price for ^{244}Cm is assumed to be $170/g. The production cost was concluded to be the same as that for ^{242}Cm and was thus set at $110/g. Although not so great as for ^{243}Am, the credit is significant and, together with that for ^{243}Am, may encourage plutonium fueling, optimization for neutron capture instead of fission, and the offering of premium prices for by-product, high-exposure plutonium with values based on ^{240}Pu and ^{242}Pu content in addition to the fuel values of ^{239}Pu and ^{241}Pu.

Plutonium Isotopes

Essentially all considerations of plutonium value have been concerned with its use as a nuclear fuel. Its possible value as the only source material for manufacture of valuable higher isotopes has been specifically avoided, subordinated, and/or discounted.[14] In view of the efforts to produce significant quantities of ^{244}Cm over the next few years and beyond, it seems unreasonable to continue to avoid consideration of credits to the value of plutonium for such production. This production will be realized with little more than an incidental investment in neutrons resulting from the normal utilization of plutonium as a fuel in thermal reactors. Through processing by conventional means, very significant quantities of by-product curium (as well as neptunium and americium) will then be recovered at costs potentially far below those now being experienced in the special production programs.

The conventional studies, which do not take into account the value of the higher isotopes, result in conservatively low values for plutonium. A similar but somewhat less significant situation exists for recycled ^{235}U fuels, if a value is placed on ^{236}U in view of its role as the precursor of ^{237}Np and eventually of valuable ^{238}Pu.

In the process to produce plutonium by irradiation of ^{238}U, all the five long-lived plutonium isotopes are produced. In irradiations of short duration, formation of ^{239}Pu is dominant. However, in power-reactor operation, in which the objective is conversion of as much low-cost ^{238}U to plutonium as possible to gain heat from the concurrent fission of such plutonium, a substantial part of the total plutonium exists as isotopes above ^{239}Pu, only one of which, ^{241}Pu, is significantly fissionable in the thermal spectrum. Heretofore, by-product plutonium has been valued only as fissionable material relative to the equivalent value of ^{235}U. This figure has been set at $10/g of ^{239}Pu plus ^{241}Pu (Table 1). No credit has been acknowledged for any of the other plutonium isotopes. Curium-244, which can be obtained only by irradiation through the whole chain of plutonium isotopes, has potential value. Therefore, as precursors of ^{244}Cm, all the plutonium isotopes, and specifically the nonfissionable ones such as ^{242}Pu, also have some value. In this study the value of the neutrons absorbed for the production of higher isotopes has been accepted as a reasonable basis for establishing the minimum value of these plutonium isotopes. The production cost of plutonium is merely the share of the fuel-processing cost (assumption 9, p. 20) borne by each gram of plutonium in a ton of 25,000 Mwd/t fuel. This figure is very close to $2/g (see assumption 10).

Consideration of the value for successively formed, higher isotopes of plutonium based on the incremental value of invested neutrons is complicated by the formation of two plutonium isotopes that have substantial fission cross sections and thus are less valuable as higher isotope precursors. In the following discussion this factor is taken into account when arriving at plutonium isotope values.

Plutonium-238. Although it exists in fuel, ^{238}Pu has no value because it cannot be separated from other plutonium isotopes. It can, however, be recovered from the curium fraction through the aging process required to obtain reasonably pure ^{244}Cm. Curium-242 credit is therefore shown only as credit for the ^{238}Pu provided by this route. The processing cost for ^{238}Pu is, then, the value of the ^{242}Cm plus the cost of separating the ^{238}Pu. The quantity of ^{238}Pu available by this route is also minor. Processing of fuel with a minimum aging period, and thus recovering curium before so much of it has decayed to ^{238}Pu (which would otherwise be "lost" in the usual mixture of plutonium isotopes), would be a means of possibly doubling the amount of ^{238}Pu that could be eventually recovered in fairly pure form. However, such ^{238}Pu would always be contaminated with appreciable ^{240}Pu derived from the decay of ever present ^{244}Cm.

Plutonium-239. Plutonium-239 at the modest value of $10/g as a fissionable material is still a very valuable material in unprocessed reactor fuel. Although it is intended for use as a power producer, additional credit may also be taken for it as a precursor of the higher isotopes. Since 26.7% of the ^{239}Pu does not fission on capturing a neutron, 26.7% of the value of the

neutrons that went into the initial formation of ^{239}Pu may be taken as credit for higher isotope formation. However, at the ^{241}Pu position another large fraction (71.5%) of captured neutrons results in fission instead of a new isotope. Therefore the value of ^{239}Pu for higher isotope production is only 7.61% (26.7 × 28.5%); neutron credit must take these value deficiencies into account. With this situation emphasized, the credit value of ^{239}Pu for higher isotope production and for fission is estimated as

$$\text{Credit} = \frac{\$ \text{ value of 1 gram atom of neutrons} \times 0.267 \times 0.285}{239} + \$10^{*}$$

$$= \frac{\$2300 \times 0.267 \times 0.285}{239} + \$10$$

$$= \$10.73/\text{g} \ (\sim \$11/\text{g})$$

The 26.7% figure is the ratio of absorption cross section to total cross section for ^{239}Pu. The 28.5% figure is the ratio of absorption cross section to total cross section for ^{241}Pu. These ratios are not fixed, and thus they may be optimized for maximum higher isotope production.

Plutonium-240. Plutonium-240 is nonfissionable in the thermal-neutron spectrum and therefore to date has not had a value set on it. It may be regarded, however, as valuable as a fertile material required for the formation of fissionable ^{241}Pu. If the value of fertile material is proportional to the cross section, ^{240}Pu would be worth about 100 times ^{238}U.† With depleted ^{238}U valued at a high figure, $4.50/lb in the form of the oxide (= $0.01/g), ^{240}Pu may be worth $1.00/g. This is regarded as sufficiently significant in this study to justify inclusion as a modest credit.

As in the case of ^{239}Pu, the neutron credit for ^{240}Pu is lowered because the successor, ^{241}Pu, is fissionable. Only 28.5% of the neutron absorptions result in formation of the next higher isotope. Therefore the neutrons absorbed to form ^{240}Pu from ^{238}U are only 28.5% effective in obtaining higher isotopes. The credit for neutron investment in ^{240}Pu should thus be

$$\text{Credit} = \frac{\$2300 \times 2 \times 0.285}{240} + \$1 = \$6.46/\text{g} \ (\sim \$6.50/\text{g})$$

Although this neutron credit is much greater than for ^{239}Pu, the total value is less because of the high fission-value credit given to ^{239}Pu.

Plutonium-241. Plutonium-241 is the other readily and thermally fissionable isotope of plutonium formed by neutron capture in ^{240}Pu. It is also valued at $10/g in a mixture with the other plutonium isotopes for use as a thermal reactor fuel. Its cross section for capture and fission in the thermal range is about the same as that of ^{239}Pu, 28.5% of the absorptions resulting in capture to form ^{242}Pu; the balance results in fission. Thus, for higher isotope production, only 28.5% of the neutrons absorbed to form it can be credited for higher isotope manufacture. The credit for neutron investment in ^{241}Pu and for fission value should thus be

$$\text{Credit} = \frac{\$2300 \times 3 \times 0.285}{241} + \$10$$

$$= \$18.16 \text{ g} \ (\sim \$18/\text{g}).$$

Plutonium-241 has a half-life of 13 years. Its decay is predominantly by beta emission to produce ^{241}Am. Plutonium that originates from fuels of prolonged exposure will be rich in ^{241}Pu, which, on storage, converts to ^{241}Am, which can be recovered in fairly simple solvent extraction or ion-exchange processes. The pure ^{241}Am thus recovered can be used as a heat source or can be irradiated to produce ^{242}Cm, an isotope of extremely high specific power (120 watts/g). As discussed earlier, ^{242}Cm on decay converts to ^{238}Pu, another isotope of even more interest as a heat or power source.

Plutonium-242. Plutonium-242 formed by neutron capture in ^{241}Pu is essentially nonfissionable in the thermal-neutron spectrum and has a very long half-life (379,000 years) and only a modest absorption cross section (23 barns). In uranium fuels of prolonged exposure or, preferably, in highly burned plutonium fuels, the ^{242}Pu content can reach very high concentrations—in some situations well over 95%. Such compositions would be ideal as target material for higher isotope manufacture—^{243}Am, ^{244}Cm, and beyond. Since it is nonfissionable, its only apparent use is for such production, and it should therefore receive full credit for neutron investment all the way from ^{238}U. Its credit on this basis is:

$$\text{Credit} = \$2300 \times 4/242 = \$38.00/\text{g}$$

*Accepted value as fissionable material.

†This is also an oversimplification since the cross sections are highly variable and dependent on specific reactor conditions.

Other Plutonium Isotopes. The *Chart of the Nuclides** shows 10 other plutonium isotopes. Except for ^{236}Pu and ^{244}Pu, they are generally of short half-lives—minutes, hours, and days—and are thus essentially without interest as practical recovery possibilities. Plutonium-236, a member of a decay chain dominated by ^{232}U and ^{228}Th, would be of considerable interest as a heat source if it could be produced cheaply, but a practical production process is not apparent. Plutonium-244 has by far the longest half-life of any plutonium isotope (76 million years). Unfortunately between it and ^{242}Pu is ^{243}Pu, an isotope with a half-life of only 5 hr. However, intense irradiation of highly concentrated ^{242}Pu in a very high flux may be a means of producing ^{244}Pu in practical quantities and in high concentrations. As mentioned under americium, ^{244}Pu also results from ^{243}Am irradiation via a small fraction of branching decays of short-lived ^{244}Am.

If quantities of ^{244}Pu of fairly high isotopic purity could be obtained, the possibility would exist for production of appreciable amounts of fairly pure ^{245}Cm. This curium isotope may be of interest as a fissionable material with a critical mass much smaller than that of ^{239}Pu, making it particularly advantageous for use in very small, compact power reactors. The route to this product through ^{244}Pu is the only apparent one, other than by isotopic separation of mixed curium isotopes.

The estimated plutonium isotope values shown in Table 4 are based on nuclide chart values for cross sections, etc. Cross sections may vary widely in a given reactor situation, and, with significant values being assigned to plutonium isotopes, conditions may be optimized for maximum production of certain isotopes. The credit values thus obtained could differ greatly from those shown in the table.

Summary of Data on Estimated Values

The economic data developed in the above discussions are summarized in Table 3. In a discussion of fission-product values, questions arise relating to the comparative prices of some of the common but costly commercial elements. Table 5 lists current prices of many such materials.

*Knolls Atomic Power Laboratory, *Chart of the Nuclides*, 9th ed., Revised to July 1966.

Table 4 Summary of Value of Plutonium Isotopes

Isotope	Use	Estimated value* per gram
^{238}Pu	Heat source	$250.00
^{239}Pu	Nuclear fuel, higher isotopes	11.00
^{240}Pu	Fertile material, higher isotopes	6.50
^{241}Pu	Nuclear fuel, higher isotopes (and ^{241}Am)	18.00
^{242}Pu	Higher isotopes	38.00

*Based on credit for neutrons that are effective in producing higher isotopes. This comment does not apply to the estimated value of ^{238}Pu.

Plutonium Recycle and Isotope Formation

Although the data on fission yields and higher isotope formation have been related almost exclusively to low-enrichment ^{235}U fuel in reactors, the important and significant impact that plutonium recycling will make on the availability of certain isotopes should also be emphasized.

Data concerning the effects of plutonium recycle on the formation of all isotopes of interest are shown in Ref. 6; information on production of the higher isotopes is also reported in Ref. 5. In view of the large number of possible fueling combinations and reactor conditions that can be conceived, only three representative situations are summarized in Table 6. The data in this table emphasize the results obtained with widely differing plutonium isotopic compositions. In view of the yield curves shown in Fig. 2, it is certain that major changes in concentrations of the fission products with masses in the region of the low-mass peak would be realized. The estimated production effects[4] under conditions of plutonium recycle in natural uranium for these fission products would be:

^{85}Kr: reduced about 50% (to 8.4 g/ton)
^{90}Sr: reduced about 50% (to 239 g/ton)
^{99}Tc: essentially unchanged (to 630 g/ton)
Rhodium: increased about 50% (to 478 g/ton)
Ruthenium: increased about 15% (to 2029 g/ton)
Palladium: increased about 70% (to 1655 g/ton)

In situations where the plutonium fuel is high in ^{241}Pu or under reactor conditions that favor the formation and fission of ^{241}Pu, the effects indicated above would be further emphasized by the expected corresponding shift in the low-mass peak. A similar situation exists in the fast fission of ^{238}U, with a

Table 5 Current Prices* of Costliest Commercial Elements†

Element	$/lb	$/g	Element	$/lb	$/g
Beryllium	54–66	0.12–0.15	Palladium	37–48 (troy oz)	1.19–1.54
Boron	25	0.06	Platinum	109–210 (troy oz)	3.50–6.76
Cesium	100–150	0.22–0.33	Plutonium	4540	10
Europium	650	1.43	Rhenium	600	1.32
Gallium	612	1.35	Rhodium	245–265 (troy oz)	7.90–8.50
Germanium	79.30	0.175	Rubidium	100	0.22
Gold	35 (troy oz)	1.12	Ruthenium	55–60 (troy oz)	1.77–1.93
Hafnium	30–175	0.07–0.39	Scandium	1362	3
Indium	2.00 (troy oz)	0.06	Selenium	4.50	0.01
Iridium	185–205 (troy oz)	5.95–6.60	Silver	2.00 (troy oz)	0.06
Lithium	9-11	0.020–0.024	Tantalum	32–52	0.07–0.11
Niobium	11–27	0.24–0.60	Tellurium	6	0.013
Osmium	300–450 (troy oz)	9.65–14.50	Thallium	7.50	0.016
			Zirconium	5.00–14.00	0.011–0.031

*From data in *Handbook of Chemistry and Physics*, 47th ed., The Chemical Rubber Co., Cleveland, Ohio, 1966–1967, and *E/MJ Engineering and Mining Journal*, **169(2)**: 22-24 (February 1968).

†Rare gases, most rare earths, and radioactive elements, except plutonium, not included.

Table 6 Plutonium Recycle and Higher Isotope Production

Isotope	Amount, g/ton at 25,000 Mwd/t		
	Low-enrichment uranium fuel	Plutonium – natural-uranium fuel*	
		Pu, composition a	Pu, composition b
^{236}U	3,169	723	685
^{237}Np	384	130	123
^{241}Am	56	271	386
^{243}Am	57	582	1,182
^{242}Cm	13	83	110
^{244}Cm	11.3	185	373
^{238}Pu	76	363	700
^{239}Pu	5,300	7,949	8,343
^{240}Pu	2,110	7,142	8,339
^{241}Pu	1,005	3,781	4,591
^{242}Pu	305	2,112	4,402
Total Pu	8,796	21,347	26,365

*Plutonium composition in feed, from Ref. 5: composition a, 1.1% ^{238}Pu, 62% ^{239}Pu, 22.8% ^{240}Pu, 11% ^{241}Pu, 3.1% ^{242}Pu; composition b, 2.1% ^{238}Pu, 48.3% ^{239}Pu, 24.8% ^{240}Pu, 15.2% ^{241}Pu, 9.6% ^{242}Pu.

significant shift of the low-mass peak to show highest yields for rhodium and palladium.

In addition to the concept of plutonium recycle, it is also possible that recycled uranium and target materials such as neptunium and ^{243}Am may be irradiated in power reactors. Under such conditions, major increases in ^{237}Np, ^{238}Pu, and ^{244}Cm production would be realized. This study has not considered these possibilities; however, they are discussed in Ref. 6.

Production of Other Isotopes

Although this study has been concerned with an estimation of the value of fission products and the higher isotopes produced in the course of normal power-reactor operation, the facilities required for the processing and recovery of these by-products would very likely be directly applicable to the processing of essentially any other isotope that may be produced in reactors. An isotope plant should have a great deal of flexibility in processing and materials-handling capability so that output could encompass a very broad array of products. Once a plant is set up to handle such active alpha emitters as ^{242}Cm, such energetic gamma emitters as ^{144}Ce and ^{106}Ru (^{106}Rh), and such combinations of alpha and neutron emitters as ^{244}Cm, modest changes or rearrangements in chemical processing facilities, such as tanks and columns with associated conventional control instrumentation, are all that is needed to ensure broadest processing capabilities. This situation encourages consideration of a greater range of products which may be accepted from the reactor operator. Some of the other isotopes that may be considered for recovery, purification, or packaging in a versatile fuel-processing isotope-recovery plant combination are given in the tabulation at the top of the next page.

Conclusions

A review of the results shown in Table 3 leads to conclusions relating to the value of fission products,

Isotope	Possible uses
Tritium	Heat, beta, and fusion-power source
^{60}Co	Heat and gamma source
^{170}Tm	Heat source
^{204}Tl	Heat source
^{210}Po	Heat and alpha source
^{228}Th	Heat and alpha source
^{231}Pa	Intermediate isotope, "new" element
^{232}U	Heat and alpha source
^{233}U	Fissile material
^{242m}Am*	Heat source, fissile material
^{244}Pu	Intermediate isotope
^{245}Cm	Fissile material

*Half-life is 152 years.

the value of neutrons, the importance of credit for effective neutrons, and the importance of credit for the higher isotopes.

In general, fission products do not appear to have great value to the reactor operator (not much basis for significant credit). This is due to the relatively low inherent value per unit, coupled with an indicated high production (processing) cost. This conclusion should not be taken to mean that fission products do not have value; it merely means that their recovery does not promise much as a basis for credit to the reactor operator or owner of the spent fuel. The fuel processor may find, however, attractive possibilities for profit among these. The realization of such profit potential could influence the bid value of fuels for reprocessing with benefit to the reactor operator. However, the pressure for increased value is expected to be small.

The existing values of ^{239}Pu and ^{241}Pu as fissionable materials with what appears to be realistic future selling prices and processing costs are significant and amount to about $63,000 or nearly 75% of the total estimated value for plutonium as projected in this study.

The value of the neutrons at $2300/g, which was used in arriving at the credit for reactor-operator investment in a number of the higher isotopes as well as the plutonium isotopes, results in high additional values. This justifies a more critical appraisal of the long-term value of the invested neutron to support or modify these credits.

The modest credits assumed for ^{243}Am and ^{244}Cm should receive attention from the standpoint of more accurately assessing their worth. In view of today's costly efforts at production of these elements, it is conceivable that the future values used in this study are far too low.

In view of the beneficial potentialities of an integrated fuel-processing and isotope-recovery facility in producing most economically a number of unique products, it appears that a fairly detailed engineering study of the nature and economics of such an arrangement and the process technology required justifies consideration. Particularly it would be of interest to establish the technology for routine recovery of the rare gases and noble metals and to determine the feasibility of routine americium and curium recovery and separation in the chromatographic ion-exchange facilities of an isotopes plant. The recovery of both americium and curium in such facilities has been firmly established.[15,16]

The assumptions on depleted uranium value and processing cost are oversimplified for convenience in this study. It was assumed that the fuel-processing cost would be shared equally by uranium and plutonium, with the value of uranium set so that it would just cover its share of the cost of processing. This value for depleted uranium is probably too high even for the future when it will be used intensively as a fertile material. However, whatever lower value is used (with a corresponding increase in processing cost shared by plutonium) would not alter the results of this study to the extent that the general conclusions would be changed.

Although there are many points of this study that may be argued, it is felt that changes would be compensating or minor. For example, the fission-product values are so conservatively low compared with the total by-product values that they could even be neglected without changing the picture insofar as credit to the reactor operator or cost of power are concerned. However, if ^{236}U (^{237}Np) and the non-fissionable plutonium isotopes were found to have no future value, the impact could be great since, of the total credit shown, about 30% is attributed to the net value of these isotopes. It is, however, felt to be unreasonable to consider these as having little or no value in view of the efforts being made today to produce these isotopes at correspondingly very high costs per gram—far greater than are shown in this study.

If it is accepted that neutrons have value that can be credited through sale of those products obtained by neutron capture, the incremental neutron value as used here may be a conservative figure. Furthermore, the net yield of the neutrons as used in this estimate is

probably high (giving low costs). Doubling the value of the neutrons (which still may be a conservative, low, basis) would add about $85,000 of total credits to the figures as listed in Table 3. Such a change would compensate for the unrealistic assumption that perhaps ^{244}Cm and ^{243}Am would have lesser value. This whole concept of substantial value or credit for by-products as they exist in the unprocessed fuel is therefore based largely on the assumed usefulness of higher isotopes plus the value of the neutrons used in the formation of these and their precursors.

It is reemphasized that this study attempts to assess conditions as they may exist in a free and competitive reactor-fuel-processing economy—with reactor operators negotiating for credit for the elements and isotopes as they exist in the unrefined and unprocessed state in the spent fuel elements. The fuel processors would be striving for improved markets and overall process economy for these products and negotiating with reactor operators or fuel owners for most favorable fuel prices and processing charges. It is, of course, unreasonable to assume that the present basis would actually, totally, and precisely exist for this complex assembly of possible products. Different concepts relating to the real value of materials, in which funds (as costly neutrons) are invested and which can only show a return after additional and perhaps prolonged and other costly processing steps are concluded, will probably have an even greater impact on the whole idea of by-product recovery and value. Such considerations could be the basis for additional studies. It is felt that the results as disclosed here form a practical basis for consideration of potential markets, prices, and credits for many of these potential by-products of an extensive nuclear power economy. (REG)

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Reactor Production and Characterization of ^{109}Cd*

By R. E. Lewis and T. A. Butler

Abstract: *Cadmium-109 produced from enriched ^{108}Cd[^{108}Cd(n,γ)^{109}Cd] had a higher specific activity than that produced from natural silver*

$$[^{107}Ag(n,\gamma)^{108m}Ag \xrightarrow{\beta^-} {}^{108}Cd(n,\gamma)^{109}Cd]$$

but, unlike the product of the silver target, contained no ^{115m}Cd. Effective neutron-capture cross sections were determined to be 1.1 and 700 barns for ^{108}Cd and ^{109}Cd, respectively.

The long half-life (453 ± 2 days)[1] and low-energy gamma radiations (0.22 and 0.88 Mev) make ^{109}Cd especially attractive for use as a low-photon-energy radiography source[2] material. In this as well as other applications, such as autoradiography[3,4] and mass, density, or thickness gaging,[5,6] high radionuclidic purity and high specific activity are desirable characteristics.

Cadmium-109 is ordinarily produced by neutron irradiation, in a reactor, of enriched ^{108}Cd targets by the ^{108}Cd$(n,\gamma)^{109}$Cd reaction, but it can also be produced from natural silver by the reactions ^{107}Ag$(n,\gamma)^{108m}$Ag $\xrightarrow[2.4\ \text{min}]{\beta^-}$ ^{108}Cd$(n,\gamma)^{109}$Cd. The purpose of the work reported here was to study the effective reactor production rates for ^{109}Cd from enriched ^{108}Cd and from normal silver targets and to characterize the resulting products with respect to activity concentration, isotopic composition, and radiochemical purity. In the course of the study, the cross section for the ^{108}Cd$(n,\gamma)^{109}$Cd reaction was reinvestigated because of wide variation in reported[7–10] values—2.8 to 23 barns—and the previously

*Based on ORNL-4247 (1968).

unreported cross section for the $^{109}Cd(n,\gamma)^{110}Cd$ reaction was determined.

Reactor Irradiations

The activity concentration of ^{109}Cd produced from ^{108}Cd enriched to about 70% was greater for a given irradiation time and neutron flux than that of ^{109}Cd produced from natural silver (Table 1). This is because the production of ^{109}Cd from ^{107}Ag requires successive neutron captures. With a reactor flux of 2.2×10^{14} to 2.7×10^{14}, the activity limits for the two products are about 2.5 and 1.5 curies per gram of cadmium, respectively. The ^{109}Cd from the silver is not carrier-free because of the $^{109}Ag(n,\gamma)^{110}Ag \xrightarrow{\beta^-} {}^{110}Cd$ reaction; the rate of production of ^{110}Cd from ^{109}Ag is greater than that of ^{108}Cd from ^{107}Ag, further decreasing the relative activity concentration of the ^{109}Cd prepared by this method. However, the cost of enriched ^{108}Cd (about $10 per milligram of 60 to 70% ^{108}Cd) compared with that of natural silver may make irradiation of silver more economical for production of multicurie quantities of ^{109}Cd. Also, the product from natural silver is preferable to that from enriched ^{108}Cd for use in radiography or in mass, density, or thickness gages because it is free from other radioactive cadmium isotopes. If ^{107}Ag enriched to about 95% should be used as a target material, a ^{109}Cd product with a specific activity of about 3 curies/g could be obtained.

Table 1 Experimental Yields of ^{109}Cd from Enriched ^{108}Cd and Natural Silver

Target weight, mg	Irradiation time,* days	^{109}Cd yield	Flux, n/(cm^2)(sec)	$\bar{\sigma}$, barns† [$^{108}Cd(n,\gamma)^{109}Cd$]
		CdO (68.6% ^{108}Cd)		
29	180	76.3	2.75×10^{14}	1.34
29	540	53.1	2.20×10^{14}	0.94
25	531	63.8	2.80×10^{14}	1.29
56	858	124.6	2.20×10^{14}	1.19
		Natural Silver		
100	160	8.6	2.08×10^{14}	0.91
100	848	89.4	2.57×10^{14}	1.24
100	848	88.6	2.57×10^{14}	1.23
			Average	1.1 ± 0.2 (S.D.)

*Represents 80% of total time in reactors.

†Effective cross sections of the $^{108}Cd(n,\gamma)^{109}Cd$ reaction calculated on the Control Data 1604-A computer with the ISOSEARCH computer code;[11] corrected for the 700-barn cross section of the $^{109}Cd(n,\gamma)^{110}Cd$ reaction.

For these experiments, targets of enriched ^{108}Cd and natural silver were irradiated in the Oak Ridge Research Reactor (ORR). The enriched ^{108}Cd had an isotopic composition of 0.6% ^{106}Cd, 68.6% ^{108}Cd, 10.4% ^{110}Cd, 5.4% ^{111}Cd, 6.5% ^{112}Cd, 2.4% ^{113}Cd, 5.1% ^{114}Cd, and 1.1% ^{116}Cd. The target materials, CdO powder and 1-mil natural silver foil, were encapsulated in quartz ampules for irradiation. The thermal-neutron flux was monitored with ^{59}Co (cross section 37 barns).

Recovery and Assay of ^{109}Cd

Recovery from Enriched Cadmium

The ^{109}Cd produced from enriched ^{108}Cd, which had an activity concentration of 2.46 curies/g, contained some radioactive ^{115m}Cd, from the $^{114}Cd(n,\gamma)^{115m}Cd$ reaction, plus small amounts of ^{60}Co, ^{65}Zn, and ^{110m}Ag. The noncadmium impurities were readily removed by sorbing all the nuclides on a Dowex 50-X8 cation-exchange column (8 mm by 15 cm) from 0.1*M* HCl solution. The cadmium was then selectively eluted with 50 ml of 0.2*M* HCl (>95% chemical recovery). The amount of ^{115m}Cd in the product depends on the amount of ^{114}Cd in the enriched target, which was 5.1% in this case, giving a product in which 12% of the activity was due to ^{115m}Cd.

Recovery from Natural Silver

The ^{109}Cd prepared from natural silver contained no radioactive cadmium other than ^{109}Cd, but curie quantities of ^{110m}Ag and ^{111}Ag were present. To remove these, the irradiated target was dissolved in 25 ml of 8*M* HNO_3, and the solution was extracted with 25 ml of 30% triisooctylphosphothionate–70% hexane, which removes >99% of the silver but <0.1% of the cadmium.[12,13] The extraction was repeated twice to remove residual silver. Then the aqueous phase was washed with hexane to remove traces of the extractant; the nitric acid was removed by evaporation with hydrochloric acid; and the HCl solution, after evaporation and adjustment to 0.1*M* HCl, was passed through the Dowex 50-X8 column to sorb the remaining nuclides. The ^{109}Cd was selectively eluted from the resin with 50 ml of 0.2*M* HCl (>95% chemical recovery). The activity concentration of the product was 1.17 curies/g.

Assay

The ^{109}Cd products were assayed using a 200-channel gamma-scintillation spectrometer with a 3- by 3-in. NaI(Tl) crystal. The millicurie content of the sample was determined by gamma-ray spectrometry using a branching ratio[1,14] of 0.039 gamma photon of 0.088 Mev energy per disintegration. The specific activity of ^{109}Cd was determined by radiochemical analysis of a known weight of cadmium.

The stable-isotope composition of the products is shown in Table 2.

Table 2 Isotope Compositions of ^{109}Cd Product from Reactor Irradiation of Enriched ^{108}Cd and Natural Silver

Mass number	Isotopic composition of radiocadmium, %	
	From natural Ag	From enriched ^{108}Cd
106	None	0.50
108	36.4	67.77
109	0.046*	0.1*
110	61.5	11.35
111	<2.0	5.42
112	<0.2	6.42
113	<0.2	<0.05
114	<0.2	7.50
116	<0.2	0.99

*Calculated from specific-activity determination.

Cross Sections

Cadmium-109

Three samples, each 1.12 mc of carrier-free ^{109}Cd [produced for this measurement by the $^{109}Ag(p,n)^{109}Cd$ reaction in the cyclotron], were irradiated 7.37 days in a reactor flux of 1.8×10^{14} n/(cm^2)(sec). The average ^{109}Cd activity at the end of the irradiation, corrected for losses by radioactive decay, was 0.912 ± 0.013 that of the starting material, indicating that the average value for the effective cross section of the $^{109}Cd(n,\gamma)^{110}Cd$ reaction is 700 ± 100 barns.

Cadmium-108

From the experimental yields of ^{109}Cd from enriched ^{108}Cd and natural silver (Table 1), the effective cross section of the $^{108}Cd(n,\gamma)^{109}Cd$ reaction was calculated to be 1.1 ± 0.2 barns. This is in fair agreement with the value 1.41 ± 0.35 barns reported by Mangal and Gill.[10] (PSB and MG)

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Section

II

Isotopes and Radiation Technology

Isotope Technology Development

Leak Detection with Radioisotope Tracers

By Martha Gerrard

Abstract: *The literature (approximately 100 references) on the use of radioisotope tracers for detecting leaks in buried pipes and cable sheaths, cross-stream units such as heat exchangers and automatic transmission systems, vehicles, water carrier and storage systems, hermetically sealed components, and miscellaneous units and systems is reviewed.*

Various methods have been devised for detecting leaks—"from soap bubbles to mass spectrometers."[1] The radioisotope-tracer method has come into use in some cases simply because it is convenient, but in others because it provides information that cannot be obtained otherwise. It is particularly useful for detecting leaks in inaccessible places. The basic principle of the technique is simple: If a radioisotope that has been introduced into a system can be detected at a spot where it ordinarily would not be, the system has a leak. The isotope used, the form in which it is introduced, and the method of detection depend on the type of system being tested.

The present review assembles in one place varied information on the techniques and radioisotopes that have been used in industrial and engineering projects for detecting leaks. The articles quoted were selected from standard reference sources.* Biological and medical studies were not considered, nor was leak detection with radioisotopes that are inherent in the system (e.g., fission-product leakage from a ruptured irradiated fuel element). Since many of the original articles have been summarized in conference papers and in early reviews,[2] there is considerable duplication in the literature. Also, some work seems to have been reported several times with slightly different slants. In the reference list here, we have stated duplication when we know that it exists, and we have indicated probable duplication by grouping references to papers of about the same date by the same authors. Internal memorandums mentioned in reviews are referenced only when they are readily available.

The articles examined are grouped below under the headings "Buried Conduits and Reservoirs," "Cross-Stream Leaks," "Vehicles," "Hermetically Sealed Units," and "Miscellaneous."

*Including *Nuclear Science Abstracts* from 1951 through 1967, *Chemical Abstracts from 1955 through 1966, and Applied Science and Technology Index* and *Engineering Index* from 1960 through 1967.

Buried Conduits and Reservoirs

Radioisotope-tracer methods are particularly useful for detecting leaks in inaccessible structures, such as buried pipes, because the radiation can be detected without direct contact of the radiation detector with the structure being tested. Some of the conduits that have been tested with radioisotope tracers are old pipelines, newly laid pipes whose trenches had to be backfilled—because of weather conditions, for example—before tightness tests could be made, and gas-filled sheaths on buried cables. The tracer selected for such tests should have a high-energy gamma emission, a low radioactivity toxicity, and a half-life comparable with the time required for the test but not so long that it will remain in the system.

Detection of leaks in inaccessible piping is extremely important industrially. For this reason, the French AEC installed at Saclay a buried pipeline in which leaks can be simulated.[2a,4a] The 250-mm-diameter stainless-steel pipes are 128 m long, have 9-mm-thick walls, and are surrounded by various soils. However, leak-detection studies have now been dis-

continued at Saclay, the procedures having been taken over by commercial firms as routine work.[4b]

Conduits for Liquids

The usefulness of the radioisotope-tracer method for detecting leaks in water mains was demonstrated by workers at Harwell[3] in a study that extended over a 2.5-year period. They used ^{24}Na as the tracer because it has a penetrating gamma emission (2.75 Mev), it is quickly prepared in high activity concentration by irradiation of 0.5-g pellets of a sodium compound, the permissible level in drinking water is high (8 μc/liter), and the half-life is short enough (15 hr) that the radioactivity introduced into the system decays before the water is discharged from the main. Both static and dynamic methods for locating leaks were investigated, with each method having certain advantages and certain disadvantages (Table 1).

Static Method. For detection of a leak in a pipe shorter[3a] than 5 km, the conduit may be filled with a solution containing ^{24}Na as sodium bicarbonate. Counters are placed outside the pipe to indicate the passage of the radioactive material. When the solution arrives at the end of the section under study, the isolating valve is closed and the pressure in the pipe is raised and held for half an hour to force radioactive solution through the leaks into the surrounding soil. Then the pressure is returned to normal, the radioactive solution is flushed out, and gamma counters, either Geiger or scintillation, are inserted in holes in the ground to detect any activity in the earth. Where leakage is indicated, groundwater samples are taken for confirmation. The smallest leak detected by this method was 2 ml/hr, and leaks of more than 100 ml/hr have been detected routinely.

A similar method was used[5] to detect leaks in central-heating piping buried in cement, which are difficult or even impossible to locate by standard leak-detection methods. In this case, ^{131}I (8-day half-life) was added to the water, and the leak was found by a detector at the surface (Fig. 1). An obvious advantage is that flooring does not have to be removed if the pipe being tested runs under a building. Because of this, sodium iodide labeled with ^{131}I was used to locate a leak in the potable waterline[6] of a church in Oak Ridge, Tenn. (Fig. 2).

Both ^{82}Br and ^{24}Na have been used in France for locating leaks in a central-heating-plant circuit.[7a,b] A mobile scintillation counter on the surface of the ground may be used to detect the activity that leaks out, and isoactivity curves plotted from the data will indicate the position of the leak. The Saclay group[7c] located a 1.5-liter/hr leak in a 35-m-long conduit (20 liters volume), 80 cm deep in the earth, with 1 mc of ^{24}Na as sodium bicarbonate. For a 40-m^3/hr leak in a total volume of 400 m^3, they used 125 mc of ^{24}Na and 300 mc of ^{82}Br in a closed circuit. They considered that 1 curie of ^{77}As as ammonium arsenate or of ^{187}W as phosphotungstic acid would be needed for the same ease of detection as 130 mc of ^{24}Na. An Austrian journal[7d] has also reported the use of $^{24}NaHCO_3$ solutions for leak testing in a heating circuit.

With much longer lengths, particularly in the operation of oil pipelines, the insertion of counters in the ground would be very tedious. Therefore the leaked activity is detected from inside the pipeline.[3b,c] Either a water or oil solution containing ^{24}Na is fed into the pipe at pumping speeds sufficient to ensure turbulent flow so that the plug of radioactive solution will cover the whole cross section of the pipe. About 1 km behind the tracer solution, a detector—a sealed container carrying two halogen-quenched Geiger counters and a slow-speed wire recorder, all battery-operated—is towed by a flanged unit similar to the impeller of a go-devil pipe scraper and is carried along the line by the flowing liquid. Points at which the radioactive solution has leaked out of the pipe are indicated by counters, whose pulses are fed to the recorder. A series of small gamma sources (each about 200 μc of ^{60}Co), placed beforehand at convenient intervals against the outer wall of the pipe, serve as distance markers. The system has been successfully used for lengths up to 20 km, the limit imposed by the recording equipment. If the pipe is not too long, the detector may be moved through the line by a cable and winch, the distance along the pipe being recorded by a cable length meter. The use of this method in Romania[8] was reviewed in an earlier issue of *Isotopes and Radiation Technology*, from which Fig. 3 is reprinted.

Small leaks in a long main may be located by the "direction method."[2a] Both the inlet and outlet stopcocks are closed, and the pressure is maintained by supplying water in the middle of the suspect section. When pressure equilibrium has been established, about 0.5 mc of ^{24}Na is injected and flows chiefly toward the largest leak. The direction of flow, and hence the direction in which the leak is located, is indicated by counters on either side of the injection point. Repeated application of the technique localizes the leak in a short section.

Besides ^{82}Br and ^{24}Na, 37-min ^{38}Cl has been used for leak testing of water pipes in Sweden.[2f]

Table 1 Characteristics of Methods for Liquid-Leak Detection*

Use	Sensitivity	Accuracy	Advantages and limitations	Radioactivity	Detection equipment
			Static Methods		
Up to 1–2 miles of any diameter pipe	Down to 0.02 gal/hr	Separation of rod holes	Cheaper than digging; restricted to leaks in predetermined positions (e.g., joints); expense of making, and difficulty in exact location of, rod holes	^{24}Na, 1–20 μc/gal	6-ft-long probes with Geiger counters incorporated, coupled to battery-operated rate meter
Particularly useful for short lengths under crossings or entrances and under standing crops; any diameter pipe	Down to 0.01 gal/hr with detector traveling 30 ft/min	1 ft	No need to know exact run of pipe; break must be made every 2000 ft; towing wire must be passed through pipe before test	^{24}Na, 1–20 μc/gal	Geiger counter in watertight container connected through 1000 ft of cable to a battery-operated rate meter
			Dynamic Methods		
Long length of constant-diameter pipe, e.g., oil distribution lines	Down to 0.1 gal/hr with detector traveling 1 mile/hr	20 ft	Up to 0 miles have been tested without access to pipe except at ends; pipe diameter must be greater than 9 in. and pumping liquid must be turbulent	^{24}Na, 1 curie; markers, 200 μc of ^{60}Co	4 Geiger counters and wire recorder, all battery operated, mounted in special container; play back to rate meter and paper recorder; portable rate meter to monitor passage of radioactive solution
Large leaks in old pipes		10 ft	Simple equipment; access needed only to outside of pipe at a few positions; leak must be large enough to give turbulent flow	^{24}Na, 500 μc	Portable battery-operated rate meter incorporating scintillation detector
Industrial and household heating supply	Variable, depending on local conditions; about 0.1 gal/hr for $^3/_4$-in.-diameter pipe	6 in.	No access required to enclosed or shallowly buried pipes except at ends; leak must produce linear flow sufficient to move ball	To monitor from ground level, dependent on depth, e.g., 10-in. cover, 50 μc ^{24}Na; 3-ft cover, 100 mc	Same

*From Ref. 2*e*.

Patents have been granted[9a] on a pipeline leak-detection method using gamma-radioactive hydrocarbon-soluble substances, e.g., $C_6H_5{}^{82}Br$, $C_6H_4{}^{82}Br_2$, $(CH_3)_3{}^{122}Sb$, and $(C_2H_5)_3{}^{76}As$, which are prepared by neutron irradiation of the nonradioactive compounds. After the tracer has been allowed to diffuse through the leak, the line is flushed and a detector is moved through it to locate the leaks. The French have also considered ^{131}I, ^{82}Br, and ^{122}Sb as well as ^{24}Na for hydrocarbon tracing.[7b] A number of labeled (^{3}H, ^{14}C, ^{82}Br, ^{131}I, ^{77}Ge, ^{75}Ge) hydrocarbon-soluble metallo-organic compounds are listed in reference 9*b*.

Another U. S. patent[10a] describes a method for leak detection in which a radioisotope tracer is injected into an oil pipeline and the accumulation of radioac-

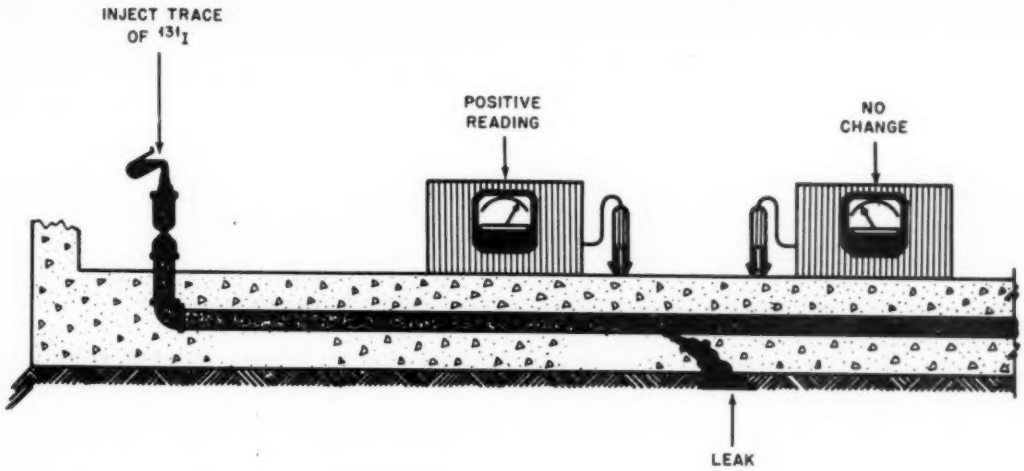

Fig. 1 Detection of a leak in concrete-buried piping by the use of ^{131}I.

Fig. 2 Detecting a leak in a church waterline, (*a*) injecting ^{131}I tracer into a rubber hose containing the flowing water, and (*b*) detecting the radiation signal through 5 in. of concrete flooring.

tivity around a leak is detected with a probe. Still another patent describes a test in which the leak is located by monitoring the spacings of the injections.[10b]

Oil-soluble ^{198}Au compounds have been developed[11] in Australia for several purposes, one of which is detection of leaks across valves in petroleum stock lines.

Dynamic Method. The velocity method of determining leaks is often used with long lengths of pipe.[3d] A small amount of high-activity material is introduced into the line, and the time required for this activity to reach a counter at a convenient access point, e.g., a hydrant, is determined. If the flow rate is high enough to produce turbulence in the liquid, e.g., 500 liters/hr in a 10-cm-diameter main, the activity may be intro-

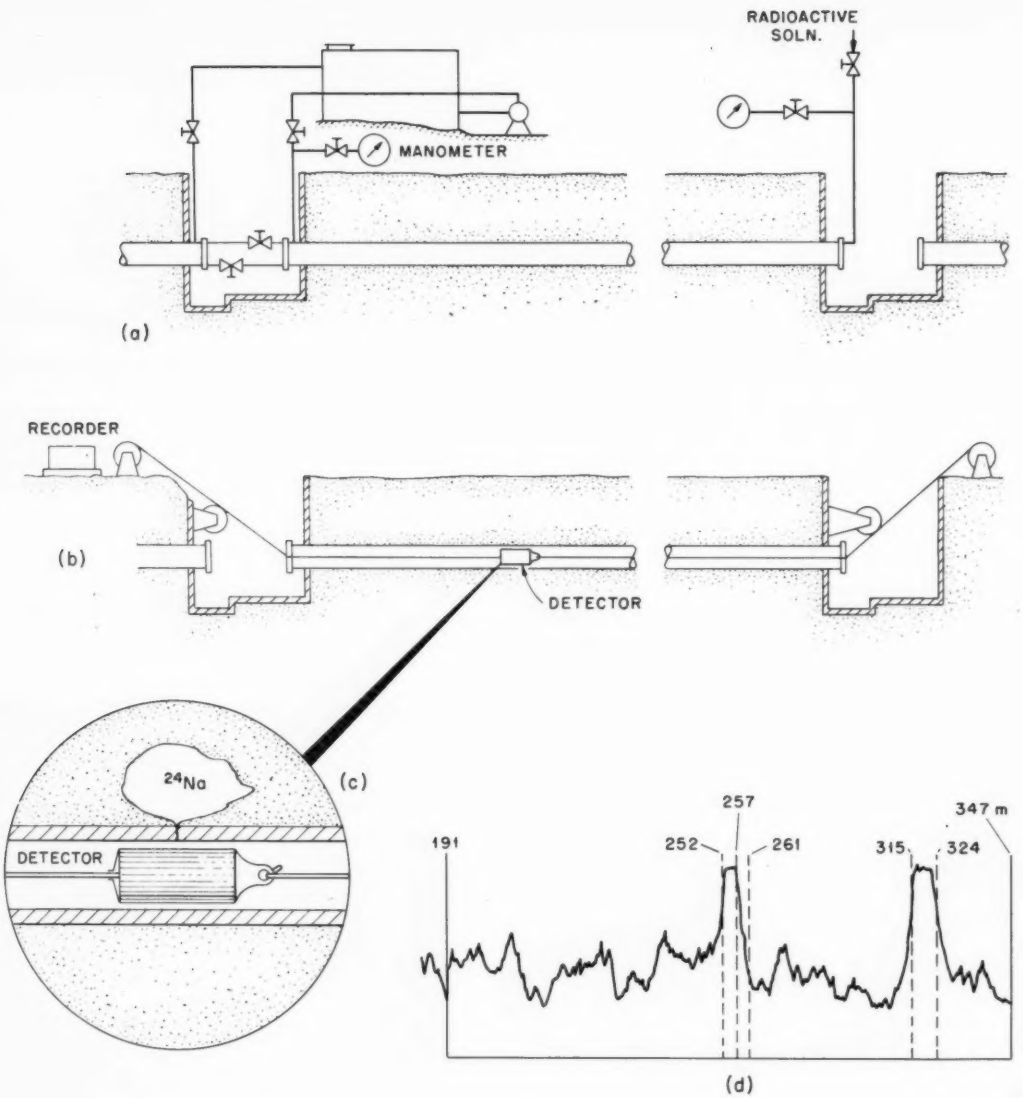

Fig. 3 System for testing leaks in pipes with radioisotopes. (*a*) Pipe through which radioactive solution is flowing. (*b*) Pipe with detectors passing through. (*c*) Enlargement at position of leak. (*d*) Record of activity showing two leaks.

duced as a solution. The velocity of the liquid changes abruptly as it passes a leak, the change being indicated by the counting data. Comparison of the apparent velocity in the section containing the leak with the velocities in the preceding and succeeding sections indicates the location of the leak within a few meters.

When the leak flow rate is insufficient to ensure turbulent conditions, the velocity of a radioactive tracer enclosed in a floating sphere can be measured.[12] A rigid sphere with projections at its equator is the best vehicle for the tracer, but a sponge-rubber ball has been used, with the tracer in the form of a wire source, for example. The float is weighted so that its density is the same as that of the fluid in the pipe. The piping course is marked out beforehand. The method is reported to detect several leaks in one system. The source stops at

the first leak if all are the same size, and a current larger than the leakage is created to sweep the float to the next leak. If the first is small and the next large, the float usually goes to the second but slows down at the first. If the first is too large for the float to be swept past, a new starting point can be chosen.

The particular isotope used in the float depends partly on the thickness through which the radiation must be detected. The 2.75-Mev gamma radiation of ^{24}Na can be easily detected even in deeply buried lines. If the float should become stuck somewhere in the pipe, the 15-hr activity would soon decay to a harmless product. For shallow lines, ^{60}Co, which emits 1.17- and 1.33-Mev gamma rays, can be used, but floats containing this isotope would have to be recovered because of its 5.2-year half-life. The amounts of ^{60}Co and ^{24}Na needed in a tracer-float leak-detection system are shown in Fig. 4 as a function of backfill thickness.

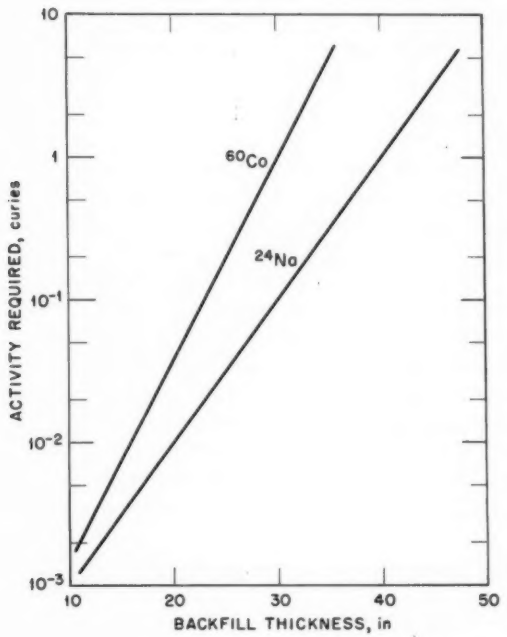

Fig. 4 Source strengths required for different backfill thicknesses.

The use of a free-flowing sealed radioisotope source to accurately locate a constriction in a hot-water pipe was demonstrated with 1 μc of ^{60}Co sorbed on a ceramic base.[13] The test pipe was up to 40 m long.

Conduits for Gas

A radioactive gas introduced into a buried, leaking gas pipe will leave the pipe through the leak and diffuse into the surrounding soil. A detector inserted into the soil, or sometimes even at the soil surface,[14] will indicate leakage of tracer radioactivity. As little as 0.2 mc of ^{222}Rn or ^{14}CO dissipated into the soil is readily detected.[15a] The amount of radioactivity that must be introduced into a buried gas pipe system in order to locate a leak may be calculated.[15b]

A frequently used tracer for buried gas mains is CH_3Br labeled with ^{82}Br. Satisfactory results were reported[16] when the tracer was injected into the buried gas main, held under pressure, and then released, with a detector on the surface of the ground to indicate where the radioactive gas had leaked from the pipes and diffused into the soil. The method was reported to be rapid, simple, and convenient and to locate leaks exactly. The labeled methyl bromide may be prepared either in the laboratory[17] or at the site.[4a] Leaks of less than 50 liters/hr have been detected with ground-level detectors.

In Australia, where 10 to 20% of the fuel gas distributed is lost, chiefly in the line from the gas main to the consumer, some 50 lines per day are tested.[18] The 1-in.-diameter individual service lines are buried about 4 ft deep and are up to 60 ft long. Leaks can be located within 6 in. About 90% of the 500 μc of $CH_3{}^{82}Br$ used per test is condensed and reused.

Optimum conditions for leak detection with ^{82}Br and ^{222}Rn have been determined and the method has been used in Poland.[19]

Another isotope suggested for use in gas pipelines is ^{137}Cs, which could be injected into a line as an aerosol plug, for example.[20]

Gas Reservoir

The nuclide ^{41}Ar was used to determine the path of leakage from an underground gas storage reservoir.[21]

Cable Sheathing

Gas-filled sheathing on buried cables may also be leak-tested with radioactive gases. For example, ^{82}Br gas injected into the middle joint of a cable flows toward a leak. As in the case of liquids, the direction of flow is indicated by G-M counters placed on both sides of the injection and the leak is localized by moving the detectors.[22]

Some of the long-distance telephone cables in France contain paper-insulated wires in a jute-covered

lead envelope under 0.8 atm nitrogen. These are buried 0.8 m below the ground surface, and any hole leads to disastrous consequences if not repaired before water leaks in. Leaks of 0.08 to 0.2 liter/min, which are considered small by the telephone people, were found by injecting $CH_3{}^{82}Br$ and allowing it to stand overnight.[23] A radioactive spot developed above the leak.

Radon has also been used[24] for locating leaks in cable sheaths containing pressurized gas. Leaks may be found to within 1 sq ft.

A U. S. patent[25] has been issued on a method for detecting a leak in a cable sheath by introducing a radioactive gas, under pressure, into the sealed inner sheath and flowing a nonradioactive gas over the sheath to pick up any leakage. Detectors are moved along the length of the installation.

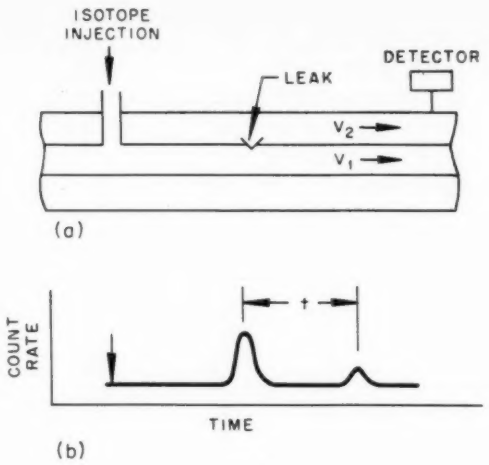

Fig. 5 Use of a radioisotope tracer to detect a leak in the inner pipe of a two-pipe concentric system: (*a*) isotope injection and detection scheme, and (*b*) radioactivity record.

Cross-Stream Leaks

A leak from the inner pipe of a concentric pipe system to the outer, as in a heat exchanger, can be found with the use of radioisotope tracers.[26] Since the detector is placed directly on the piping, isotopes emitting low-energy radiation may be used. The isotope may be retained in the system to permit decay, and safety considerations do not require that the half-life be inconveniently short.

In this technique a small amount of radioisotope is injected into the central pipe, where the isotope is confined until it arrives at the leak (Fig. 5*a*). Here some of the tracer goes into the outer stream and travels at a different velocity so that a detector located outside the pipe, downstream from the leak, indicates two pulses (Fig. 5*b*). The distance of the leak from the detector is given by $V_1V_2/(V_1 - V_2)t$, where V_1 and V_2 are the velocities in the inner and outer sections of the pipe, respectively, and t is the time between the centroids of the recorded pulses. Leaks in a 40-m-long line, where the diameters of the inner and outer pipes are 10 and 15 cm, respectively, have been located to within a few inches.

When the inner pipe contains a process stream that is heated or cooled by a medium flowing in the outer pipe, impurities in the product may indicate a leak from the outer pipe into the process line. If there is such a leak, a radioisotope tracer injected into the coolant at its inlet will be detected by a G-M counter at the process line exit (Fig. 6). However, detection alone of such leaks may not be sufficient. Small leaks may be tolerable, and costly repairs to the heat-exchanger unit may accomplish little if the bulk of the impurities are coming from elsewhere in the line. In this case the size of a leak should be determined,[27a] e.g., by the

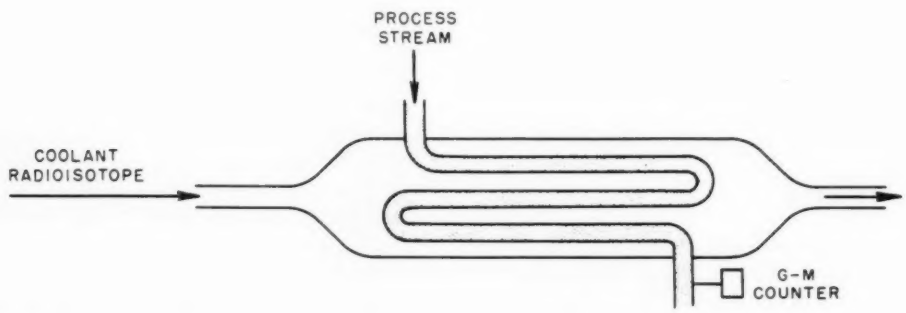

Fig. 6 Method for detecting leakage from cooling medium into a process stream.

total-count method.[27b] In this method the amount of tracer, A, that leaks into a line is found from $A = NQ/F$, where N is the total counts leaking in, Q is the leakage rate, and F is a system constant. The method was applied in a refinery[27c] where excessive water in a gasoline line was thought to be due to a leak from the 75-gal/min water stream that cooled the line. However, when a cesium chloride solution containing 2.3 mc of ^{134}Cs was injected into the cooling water, only 300 ± 85 counts of activity appeared in the gasoline line, where water was accumulating at a rate of 1.4 gal/min. Thus the activity inleakage was 0.9 μc (i.e., 300 × 1.4/480, where $F = 480$ for the system), which is only 0.4% of the 2300 μc injected. From this it follows that, of the 1.4 gal/min of water leaking into the gasoline, only 0.4 × 75 = 0.03 gal/min came from the cooling water. Therefore other leaks into the gasoline line had to be sought.

In a synthetic ammonia plant,[28a,b] where the gas flow in the heat exchanger is 3 million scf/hr, tests with 0.3 to 0.5 μc of ^{85}Kr showed that less than one one-hundredth of a 0.5 to 2% loss of synthesis gas was due to an exchanger leak. Thus repairs to the exchanger would not have controlled the loss. With flow rates this large, where even a 1% leak would require a large amount of tracer, it is more economical to use the "total-sample" method,[28c] a variation of the "total-count" method. Here a sample is collected continuously from the traced stream and counted later with a more efficient counter than can be used when the counting is done in situ.

In another heat-exchanger test,[28b] 1 curie of tritiated water was used. The test was started in the afternoon, results were reported later the same day, and the plant was ready to start up on time the next morning.

In a cellulose acetate process system, a leak of up to 1 liter/hr from the cooling circuit into a 5-m^3/hr cellulose acetate line was detected with 15 mc of ^{24}Na injected into the water coolant.[7b]

A leak of the order of 0.1 liter/day from the cooling-water side (5000 m^3) of a steam condenser to the condensate well containing 5 m^3 of water was detected with radioactive ammonium bromide.[2f]

A simple, direct method for use in a garage was needed to determine whether, in an automatic transmission system, fluid was leaking from one section to the other. A method was developed using such small amounts of ^{124}Sb that no radioisotope usage license was required.[29]

A U. S. patent[30] has been granted on a method for detecting and measuring leaks in which a radioactive-tracer compound is injected into the feed and a sample of the product is removed downstream from the heat exchanger. The chemical properties of the tracer are such that, if there is a leak in the heat exchanger, some of the tracer will bypass the reactor and show up in the effluent as unconverted product.

In an underground system where coal is burned in the seam and the hot gases are returned to the surface for use as a source of heat, air is carried to the burning zone by a network of pipes. A leak between coaxial pipes in such a system, 130 ft deep, under turbulent conditions was determined (Fig. 7) by injecting 100 mc of ^{85}Kr into the airstream as a short pulse.[31] The time required for the pulse to return to the surface and the

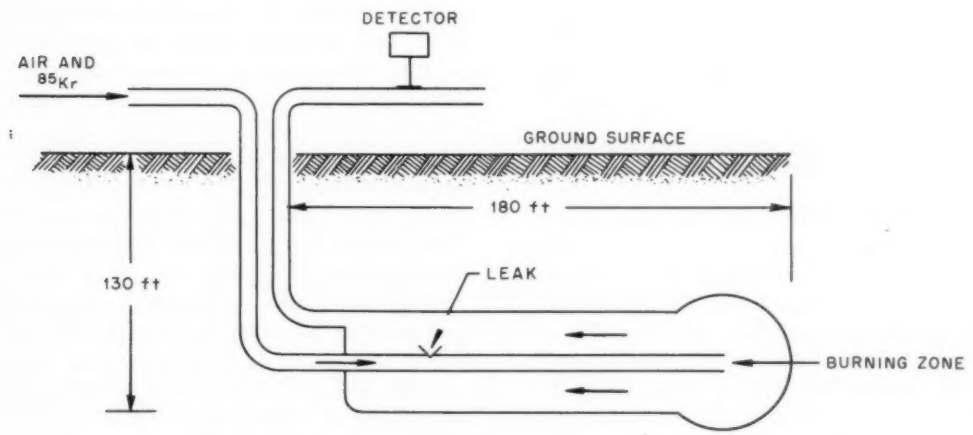

Fig. 7 Detection of leak in air-supply line to underground coal-burning system.

characteristics of the returned pulse indicated the location and size of the leak.

The leakage of cooling gas from the primary circuit to the secondary in a nuclear power plant steam generator was measured[31b] with ^{41}Ar.

Calculations[32] have indicated that upstream diffusion is not a serious problem when a radioactive gas leaks into an airstream.

Vehicles

The use of ^{85}Kr for detecting leaks in the Saturn space vehicle has been under study for some time.[33] Krypton-85 was selected for this use because it is efficient, safe, economical, readily available in large quantities, easy to detect in low concentrations, and soluble in space vehicle fuels. The vessel to be tested is filled with a $N_2-^{85}Kr$ mixture under pressure, and the leak is located by moving a detector slowly along the outer surface of the vessel or line. A leak can be detected in a few seconds. Both G-M tubes and solid-state detectors have been used.

Tritium has also been studied for this use.[34]

For determining[35] where CO from an airplane engine was leaking into the cabin of an airplane, ^{85}Kr was admitted to the intake manifold at a rate of 150 to 200 μc/min. Detectors were placed at various points in the cabin, both on the ground and in flight, and the leak point was found. Detection would have been very difficult, if not impossible, by other methods.

For detecting leaks in railway cars,[36] which can interfere with proper ventilation, ^{85}Kr was concluded to be better than ^{133}Xe. Tests were made in a test chamber large enough to hold an entire car and designed to simulate conditions of a train moving at speeds up to 120 km/hr with extremes of temperature and humidity apt to be encountered.

A formula for calculating a leak rate from data obtained with ^{85}Kr was published in East Germany.[37]

Water Carrier and Storage Systems

Seepage and leaks from canals, reservoirs, or storage tanks may result in serious water losses or in collection of excessive water on adjacent lands. Data on such losses, for use in control, have been determined with several radioisotope tracers.

Tritium (0.0186-Mev beta, 12.26 years) as HTO, which is not appreciably sorbed on soil but migrates essentially the same as ordinary water, was used in studying[38] seepage from an 800-ft section of the Madera Canal in California. Into the 6710-m^3 volume of water, 10 curies of HTO was injected to give a concentration of 1.5×10^{-3} μc/ml after thorough mixing. Tritium concentrations in test wells were determined at intervals of up to 30 days in order to show the rate and direction of the leakage. From laboratory and other data, the experimenters concluded that ^{3}H is preferable to radioactive chlorine as a tracer for this application. The use of $NH_4{}^{82}Br$ to determine the leakage rate from a canal bringing water from a lake to a small hydroelectric plant is mentioned in a Swedish report.[2f]

Seepage from a reservoir in Wyoming was determined[39] by injecting ^{3}H into the reservoir and determining the tracer concentration in 1000-ft-deep drill holes around the reservoir.

Using ^{131}I, French workers[40] established the cause of water losses from the reservoir behind an earth dam. Iodine-131 was injected at points along a 50-m stretch. The radioactivity of the water at drainage points was determined, and, from the data, curves were drawn to show where the radioactive "cloud" reached bottom in the reservoir. Several modifications of the method to suit special situations were given by these authors.

A 70-mc pulse of ^{131}I was used[41] in Japan to determine leakage under a dam. The injection was made upstream of the dam, and samples taken at a well on one bank and at springs in the riverbed were analyzed for ^{131}I to show the amount of leakage (Fig. 8).

Both ^{131}I and ^{82}Br were used to find the rate of filtration of groundwater through a filter gage tube.[42] From the data the seepage paths in the curtain under a dam were determined. In a location where calcium hydroxide solution was being injected under a dam to absorb CO_2 in leakage water and so protect soluble calcite in downstream rock zones, ^{24}Na was added to the stream to determine whether the CO_2 was continuing to leak through.[2f] In Korea,[43] geological data indicated a leak in a dam to be about 80 m away from the point of intersection of the dam with a mountain near the shoreline. Data obtained with 10 to 15 mc of ^{24}Na, as NaCl, located the leak only 52 m away from the intersection.

Polish workers,[44] reporting on a seepage investigation extending over several years and with tests on a number of dams, were enthusiastic about the results obtained with radioisotopes. Their studies, which were qualitative, included work with ^{51}Cr as Na_2CrO_4, ^{82}Br as KBr and NH_4Br, ^{131}I as NaI and KI, ^{124}Sb as $SbCl_3$, and ^{198}Au as $HAuCl_4$.

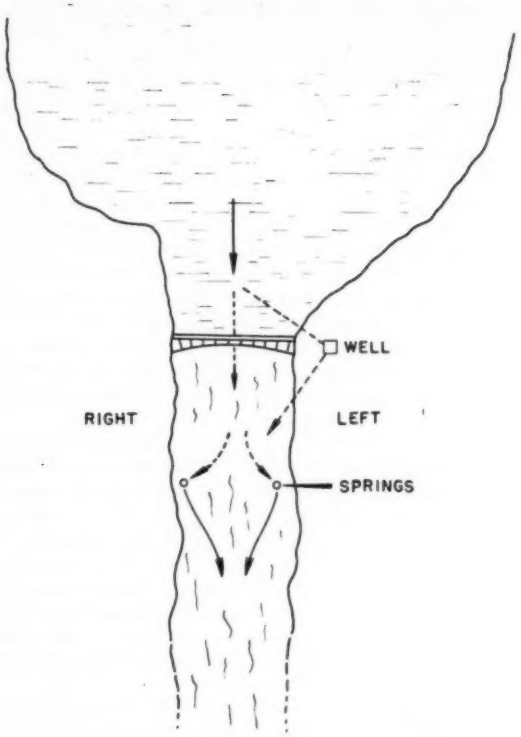

Fig. 8 Determination of seepage under a dam using ^{131}I. ---underground flow; ——surface flow.

The use of radioactive iridium complexed with ethylenediaminetetraacetate (EDTA) for detecting a leak in a reservoir was suggested by a hydrology panel meeting in Vienna.[45] However, they also felt that better radioactive tracers should be sought, e.g., EDTA-complexed Cr, Co, Mn, or Ru, suspended carbon black, or suspended plastic.

Hermetically Sealed Units

Leaks as small as 10^{-13} standard ml/sec in hermetically sealed containers may be detected, under favorable conditions, by the radioisotope-tracer method.[46] The method is cheaper, simpler, more reliable, and sometimes more sensitive than the use of helium with a mass spectrometer.[47] One method for this has been named the "Radioflo" method.[48]

There are two general methods[47] for carrying out this procedure. In one, the container, e.g., a transistor capsule, is immersed in ^{85}Kr at high pressure for a given time, and the capsule is monitored for radioactivity after removal from the radioactive gas. Care must be taken to remove all surface contamination of the unit before monitoring. To detect a leak of 10^{-12} standard ml/sec in a thermionic valve, it was necessary to immerse the unit for 3 days at 2 atm pressure. A large number of small-volume containers are easily tested this way. For small numbers of large containers, such as pressurized vessels and reactor fuel elements, a small amount of gas may be injected into the unit before the final sealing. After being sealed, the unit is placed in an evacuated chamber, and any leakage gas is collected and counted for radioactivity. The use of ^{85}Kr for detecting leaks in hermetically sealed components has also been reported by several other authors.[49]

The radioisotope-tracer method was used to test, prior to installation, the repeaters for the cable between France and North Africa.[23a] These units, which receive the signals from one line and transmit them to another, are enclosed in polyethylene hose filled with liquid silicone and are placed in the cable at intervals of many kilometers. Pressure on the cable may reach 400 atm, and a slow leak in the repeater shell would make the unit useless. The units were immersed in a solution of radioactive sodium carbonate under 400 atm pressure for 5 hr. Only one unit showed activity after the test, and it had solution inside it. The others were installed and worked satisfactorily.

Krypton-85 has been used in testing the sheaths of nuclear reactor fuel elements for leaks.[47c,50] In one patented method,[51] an amount of ^{85}Kr clathrate[53] is placed inside the sheath, and after the sheath has been sealed, the fuel element is heated to release the ^{85}Kr, which will diffuse through any leaks. In another patented method[52,54] for testing fuel elements, a gas containing a radioisotope tracer is placed in the free space in the end of the fuel element prior to sealing the sheath.

Miscellaneous Techniques

In the literature search, several radioisotope-tracer leak-detection procedures were found which either do not fit readily into the above classifications or are very general. These are mentioned here since they may suggest a use to a reader.

Leaks in large bitumen-lined storage tanks have been detected by placing a ^{24}Na solution in the lower part of the tank, removing the solution after enough time has passed for the solution to pass through the leak, and examining the inside of the tank with a portable scintillation counter.[2b] To detect leaks in

lead-lined wooden tanks, an oil solution of radioiodine was placed in the tank and then withdrawn and the residual activity measured.[55]

Internal leakage in a mixed-phase refinery unit was measured[56] with 7 mc of ^{82}Br-labeled bromobenzene to show the bypassing of the catalyst bed in a naphtha desulfurizing unit. The minimum detectable bypassing was 0.05% of the flow through the unit.

A method for detecting areas of drilling-fluid loss and casing leaks in bore holes involves using drilling fluids of various particle sizes, each size associated with a different radioactive tracer. The sizes are chosen so that selective seepage will occur at leak points and the tracer will identify the leak.[57]

A Swedish patent has been issued for a leak-detection method in which the material in a pipeline is made radioactive and a detector is passed through an adjacent channel.[58]

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Present Status of Activation-Analysis Applications in Criminalistics*

By R. E. Jervis†

Abstract: *The application of neutron activation to crime investigation is discussed; the areas covered are: (1) Toxicology—detection of arsenic, mercury, and thallium in hair; phosphorus in the liver; selenium in blood; chlorine- and bromine-labeled toxic pesticide residues in grains; and (2) Transference of Forensic Materials—analysis of ballistic residues on skin and clothing and identification of bullet holes; identification of soil transferred from the scene of a crime; activation comparisons of human hair; analyses of fibers and tissues; characterization of drugs; activation matching of physical evidence such as glasses, bullets, and paints. The author also evaluates the importance of the technique.*

Recently a considerable interest has arisen in applications of nuclear radioactivation techniques to crime investigation. Today in criminalistics, i.e., the scientific investigation of crime, new analytical methods are being sought which will permit a wider scope for undertaking the unique and specialized analytical demands presented in each crime—often because of the scarcity or minuteness of evidence and the apparently antithetical need to establish highly reliable conclusions to be drawn concerning the link between the evidence of a crime and the suspect.

There are a number of features of the activation method which make it a valuable addition to the analytical tools already available to the criminalist: very small traces of material which might establish that a poisoning has occurred or might link a suspect to a crime are susceptible to the sensitive activation analysis even if minute specks of evidence are the only evidence available; detailed analyses of various trace constituents may serve to characterize the material as to source or allow comparison with specimens from the suspect or scene of the crime; some activation-analysis techniques may be applied without appreciably altering the physical or chemical state of the specimen so that it may be preserved for further analyses or for court exhibit. Possibly the most important feature of the technique as a specialized method for application to forensic materials is that it permits interference-free

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analyses at the nanogram level for most elements and in almost any type of matrix.

In any crime in which there is some transfer of material as a result of the discharge of a weapon or of violent physical contact between people and the immediate surroundings, it should be feasible to establish these circumstances from the physical remnants of the scene—provided that sufficiently sensitive methods of analysis are available. Each specimen of matter probably contains most of its chemical elements in unique proportions and could be differentiated from all others as to particular source, past history, sequence of handling or transfer operations, etc., if analyses could reveal the changes in microcomposition resulting from these events. With methods such as activation analysis, it would seem entirely feasible to attempt such investigations into the "uniqueness" of individual specimens; by way of example, applications are cited below in which more than 25 trace constituents of a material were determined and used to distinguish it from others. Thus it would appear that there is a great potential for future applications of forensic activation analysis (FAA).

Previous reviews of new activation-analysis applications and techniques in criminology have been made by Jervis et al.,[1-3] Guinn,[4-6] Gibbons,[7] and Smith.[8] Recently the First International Conference on Forensic Activation Analysis in San Diego (1966) dealt with the various technical, forensic, and jurisprudential aspects of the field, and the published proceedings of the meeting contain a number of excellent papers and reveal several significant new developments.[8]

This paper outlines the present status of forensic activation analysis with reference to applications which have been reported to date and those which appear feasible and highly promising on the basis of preliminary investigations and also indicates the general acceptability of the technique to courts of law.

The applications of FAA to date may be classified as:

1. Applications involving trace analysis in small samples: determination of traces of toxic materials deposited in or excreted from human tissues or detectable in foodstuffs or in the environment.

2. Applications involving detection of small residues of transferred material: detection of gunpowder, ballistic material, soil, etc., transferred from a weapon to the criminal or to tissue, bone or cloth near a wound, or transferred from the criminal or his effects to the victim or the immediate surroundings.

3. Applications involving multielement analyses in small samples: the characterization of different specimens of a class of similar materials such as paint, glass, narcotic drugs, fiber, or human head hair by multielement activation analyses, thereby revealing patterns of trace constituents which were incorporated as a result of the particular source or past history of the specimen.

The application of activation techniques to some of these materials has been developed to such an extent and has been supported by a sufficient body of research with typical specimens as to justify use and to gain acceptance in the law courts of several countries, namely, Canada, the United States, Israel, Australia, the United Kingdom, and some European countries. Other applications that appear feasible are currently being studied, and some time may elapse before the practical value of their use to forensic scientists can be ascertained. A number of forensic laboratories have established FAA groups or a working liaison with qualified scientists in a nearby nuclear institute or reactor center for applying the technique in actual criminal investigations on a semiroutine basis.

Applications of FAA to Toxicology

Arsenic Analyses in Tissues

Possibly the first documented use of activation analysis for toxicological purposes and its introduction in evidence was by Griffon[9] of Paris in the reputed arsenic poisoning of L. Besnard. The victim's hair was found to contain abnormal quantities of arsenic in an amount nearly 50 times as great as normal. An attempt was also made in this analysis to examine the distribution of arsenic along the length of the hair simply by scanning the irradiated hairs with a Geiger counter through a slit in a lead sheet. A general decrease in radioactivity with increasing distance along the hair from the root end was found; in addition, several regions of 10 to 15% greater relative intensity near the growing end were attributed to arsenic deposition and were reported to have decayed approximately with the 27-hr ^{76}As half-life. These results were surprising since the bulk of the radioactivity in neutron-irradiated human hair is due to ^{24}Na and ^{82}Br, and the relative contribution from ^{76}As is less than 1% even at increased concentration levels. These analyses were seriously questioned, but later the presence of abnormal amounts of arsenic in the Besnard hair was confirmed by Joliot-Curie.[10]

Simultaneously Smales and Pate[11] developed a radiochemical procedure for use in the activation analysis of arsenic in biological tissues and demon-

strated increased amounts of arsenic in the hair and nails of persons who had been exposed to arsenic. The method consisted in the distillation and precipitation of arsenic to separate it from large amounts of interfering sodium, bromine, and other nuclides in materials such as hair, nails, and urine from toxicological investigations.

Radiochemical procedures for the neutron activation determination of arsenic deposited in the hair and nails of both healthy individuals and the supposed victims of criminal poisoning were further developed by Jervis[12] and by Mackintosh and Jervis.[13] The radiochemical separation of ^{76}As from irradiated tissues was based on the extraction of arsenic triiodide into chloroform,[13] since it was found that greater decontamination and higher recovery could be obtained than with the distillation procedure of Smales and Pate.[11] Normal levels of arsenic in healthy persons were found to lie in the range 0.5 to 1 ppm in these tissues (a range found also by Smales and Pate), while the ingestion of abnormal (i.e., milligram and greater) quantities of arsenic was indicated by levels increased to well above 2 ppm. In this work the possibility of increased levels of arsenic deposition in the growing root ends of hair was investigated by sectioning bundles of hair into short lengths, corresponding to a few weeks' growth each, for separate analyses.[12]

The most extensive series of arsenic activation analyses in human hair effected by specific radiochemical separation from 1000 irradiated specimens was reported by Lenihan and Smith[14,15] and indicated that contents of 0.5 to 1.5 ppm in men and 0.1 to 1 ppm in women were common while contents greater than 2.5 ppm rarely occurred. These conclusions were supported by arsenic analyses of head hair and beard clippings from individuals who had inhaled toxic quantities of arsenical fumes in which arsenic contents greater than 10 ppm were found. In the same laboratory a few hair strands identified as relics of Napoleon taken at the time of his death were found by neutron activation to contain greater than 10 ppm arsenic and judged to be indicative of arsenic ingestion before death.[16,17] Activation analysis for arsenic (and also for mercury) was also carried out on the 400-year-old remains of King Erik of Sweden to confirm suspicion of a criminal poisoning.[18]

Krishnan and Erickson[19] reported an efficient ion-exchange procedure for use in activation determination of arsenic in biological materials of forensic importance and for application to toxicological investigation in suspected criminal poisonings.

A large-scale survey of trace elements in human hair by instrumental, nondestructive activation analysis (described in detail below) was completed by Perkons and Jervis.[20,21] The arsenic content of hair from about 800 persons was found to lie in the range 0.2 to 3 ppm with a modal content of about 1.5 ppm. These results are presented in histogram form in Fig. 1 and take the shape of a skewed distribution similar to that found also by Lenihan and Smith.[14,15] Thus it is certain that appreciably higher arsenic concentrations may be judged to be strong evidence of the ingestion of abnormal quantities of the element.

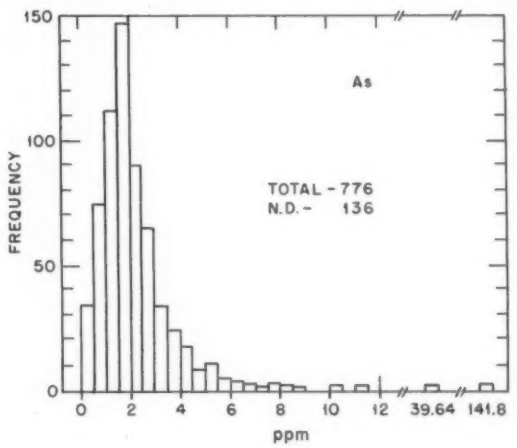

Fig. 1 Hair arsenic-content distribution in the population.

The reliability of activation analyses for arsenic in toxicological investigations has been examined by Olivier,[22] Lima, Atalla, and Silva,[23,24] Gomez and Baro,[25] Smith,[26] and Shapiro,[27] with arsenic deposition in hair through external absorption of sweat or capillary uptake within preformed hair considered as possible alternatives to internal deposition during growth.

Notwithstanding, the technique has been used and introduced as definitive evidence in high courts in homicides by Smythe[28] and by Erickson.[29] In the latter work the arsenic distribution along the length of hair bundles revealed significant concentration increases at times corresponding to periods of acute gastric distress and hospitalization leading up to death. The results are presented in Fig. 2 for different sections of hair taken from the deceased.

Concentrations in excess of 300 ppm were reported in the root ends of hair taken at the time of death, and

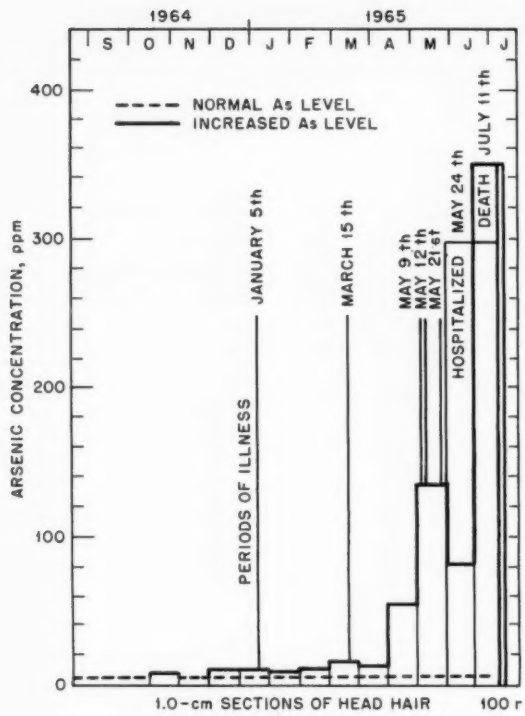

Fig. 2 Distribution of arsenic along hair from victim of criminal poisoning.

this level is nearly 1000 times as high as is normal for female head hair. Supporting research with guinea pigs given acute and subacute doses of soluble arsenic has revealed that arsenic is not primarily incorporated into the hair from sweat but does concentrate along the length of hair as a direct function of the time elapsed after ingestion.[29]

Mercury

Mercury is a toxic element whose determination in tissues and foodstuffs by neutron activation is quite sensitive. Trace mercury analysis is not only difficult but also very necessary since the WHO–FAO-recommended safe upper limit for mercury content in foodstuffs is 0.05 ppm. Sjorstrand, Christell, et al.[30-32] determined mercury in biological materials by activation of the 65-hr ^{197}Hg, whereas Perkons and Jervis,[20] Kim and Silverman,[33,34] Smith,[35] Ukita,[38] Leddicotte and Reynolds,[36] and others have reported determination by means of the long-lived ^{203}Hg nuclide. Ukita and Ohuchi[37,38] have found concentrations of mercury in excess of 100 ppm in Japanese persons exposed to soluble mercury in their diets. In cases in which the mercury ingestion was not fatal, the decrease in deposited mercury in the hair with time was explored by sectioning bundles of hair and determining the variation in mercury along the length. Head hair from a number of healthy Japanese controls was found, both by dithizone colorimetric analysis and by activation analysis, to contain from 6 to 10 ppm mercury. However, the former method tended to yield consistently lower results, suggesting either an interference in the determination or an incomplete liberation of mercury from the organic-bound state in such biological material. The mercury content in the hair of many healthy persons was determined in the large-scale hair study (see below) by Perkons and Jervis.[20] The mercury content of head hair obtained mainly from persons residing in North America was found to lie from 0.2 to 3 ppm, with a modal content of 1.7 ppm; this was appreciably lower (by a factor of 5 to 6) than average Japanese hair, indicating that the mercury intake through diet and other environmental sources is much less in America than in Japan. A similar occurrence of increased mercury in hair has been reported recently in Sweden by Westermark and Sjorstrand.[39]

Smith[8] reported forensic investigations of mercury poisonings by activation techniques in hair, skin, and nails of dentists and dental technicians.

Thallium in Hair

The concentration of thallium in human head hair in a case of thallium poisoning was observed by Jervis and Kolosta.[40] The nuclide ^{204}Tl was radiochemically separated from neutron-irradiated hair and a concentration of 0.3 ppm found, whereas, in hair from healthy persons, no thallium concentrations greater than 50 ppb could be detected. A similar analysis was performed in Germany and an unsuccessful attempt made to introduce the result in court as evidence of an alleged criminal thallium poisoning.[41]

Toxic Phosphide

By combining preirradiation chemistry with neutron activation, Krishnan[42] was able to extract abnormal quantities of a toxic phosphorus-containing compound from the liver of a deceased person. The technique was sensitive down to 0.02 μg. The absence of appreciable interference from sulfur-containing materials (i.e., from fast neutron activation of sulfur) producing the same ^{32}P nuclide was also demonstrated in this work.

Determination of Other Toxic Elements

Selenium is another toxic element, and activation analysis was used by Tomlinson and Dickson[43] and by Perkons and Jervis[20] to determine tissue and blood contents in both healthy persons and those who had been exposed to abnormal amounts.

Guinn and Schmitt[44] have reported the detection of toxic chlorine- and bromine-containing pesticide residues in illegally distributed treated cereal grains.

Value of Activation Analysis in Toxicology

Activation analysis is useful in toxicological analyses for inorganic or metallic poisons at very low concentration levels or when the size of sample available for analysis is necessarily limited, such as biopsy specimens and external tissues such as hair, nails, or skin. In order to judge whether abnormal quantities of some toxic element have been ingested, it is necessary to compare results with statistically significant data on the concentration of the element in similar tissue from a large number of randomly selected "normal" or healthy persons; few such data are yet available in the literature except those provided by a few trace-element surveys in certain tissues such as hair[20,62] and in blood.[45] Some caution should be exercised in the interpretation of the toxicological analysis of tissues or fluids in cases of suspected criminal poisonings unless well-selected controls are also studied. Circumstances of occupational or industrial exposure to pollutants can lead to anomalously high body levels, as can unusual medical practices of previous periods of history. For example, the meaning of high arsenic content in Napoleon's hair at time of death[16,17] is not clear since various potions were often administered in those days as remedies. This caution is particularly valid when the trace tissue levels involved are very low (i.e., ppb to ppm levels) and methods such as activation analysis are used to reveal increased levels.

Transference of Forensic Materials

Analysis of Ballistic Residues on Skin and Clothing

A method for the detection of gunpowder residues on the hand or cheek of a person who had recently discharged a weapon was reported by Ruch, Guinn, and Pinker.[46] Submicrogram quantities of antimony and barium contained in cartridge primer were determined by neutron activation of material removed from the hand of a suspect by a paraffin casting procedure and followed by radiochemical isolation from the irradiated material. It was demonstrated that, although a trace of antimony and barium (e.g., ~0.01 μg of antimony and 0.1 μg of barium) could often be detected on a normal hand, an appreciable increase occurred through the firing of a weapon (e.g., 0.2 to 0.5 μg of antimony and 0.5 to 1.5 μg of barium per single firing of a .38-caliber revolver). Typical results are given in Table 1. Most of this material was removed on normal washing of hands, so that valid results could be obtained only if the procedure could be applied between the time of discharging a firearm and subsequent washing of the skin.

Table 1 NAA Determination of Gunshot Residues on Firing Hand[47]

(Single Firing, .38-Caliber Revolver)

Barium, μg		Antimony, μg	
1.03	0.58	0.22	0.17
0.99	0.37	0.27	0.19
0.40	0.32	0.12	0.26
0.27	0.38	0.18	0.22
0.82	0.54	0.28	0.26

Mean result: 0.57 ± 0.27 μg Ba; 0.22 ± 0.05 μg Sb.
Hand blank (no firing, either hand sampled): 0.13 ± 0.11 μg Ba; 0.015 ± 0.012 μg Sb.

Analysis of transferred gunshot materials from the barrel of the weapon to the clothing of a victim or adjacent surfaces has been undertaken in several laboratories. Baumgaertner, Staerk, and Schoentag[48] used neutron activation and radiochemical separations to detect traces of antimony (from 1 to 124 x 10^{-10} g Sb/cm^2) on surfaces placed from 1 to 3 m distance from the muzzle of a .38-caliber pistol and in an area between 1.5 and 5.8 cm from the bullet hole. Simultaneously Krishnan[49] developed an activation procedure for determining the distance (over a range from a few feet up to 15 ft) of a firearm from a collecting surface of skin, clothing, or filter paper, based on the radiochemical separation and determination of submicrogram amounts of copper and antimony. The antimony deposition was found to be much less in firing copper-clad bullets than in firing unclad lead bullets. These results suggest that the antimony collected on surfaces surrounding the bullet entry area

originates from the antimony hardener component added to the lead bullet rather than from the antimony traces in the ammunition primer which deposit on the firing hand. It was shown that these gunshot materials remain on fabrics even after handling, washing, or dry cleaning. Since the amounts of material vary with the weapon and type of ammunition, a calibration of quantity as a function of distance must be performed. Using these techniques, a series of distance determinations, accurate to a few inches, can be made; criminal shootings at 3 to 15 ft distance can be clearly distinguished from accidental discharges or self-inflicted wounds originating within arm's reach (less than 3 ft). The variation in antimony and copper deposited per unit area on surfaces at varying distance from a firearm is shown in Fig. 3 and is given by the equation:

$$(\mathrm{Sb}) = A(FD)^m$$

where FD is firing distance and A and m are parameters for each weapon and ammunition type.

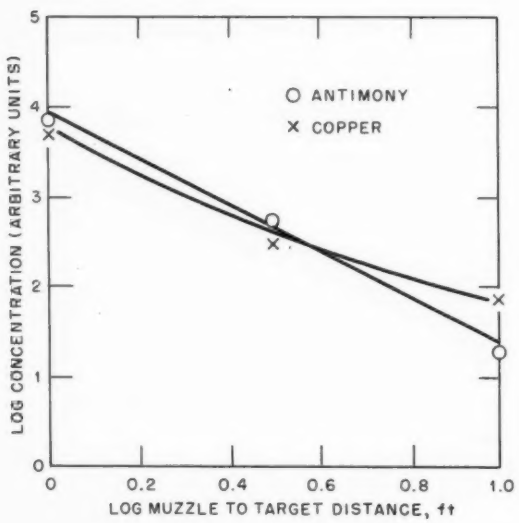

Fig. 3 Variation of antimony and copper residue with firing distance (Magnum Ruger revolver; CIL ammunition).

Since antimony (and to some extent copper) is deposited in varying amount on the shoulders or fringes of the bullet entry hole as a result of a "wiping" action of the bullet, this area is omitted in determination of distance by activation analysis of deposited antimony.

Identification of Bullet Holes by FAA

Bullet material transferred to leather shoes and also to paper and cloth at the impact site was reported by Schlesinger, Pro, and Hoffman[51] even at firing distances of 50 ft and greater, by nondestructive techniques.

Erickson, Krishnan, and Perkons[52] reported the detection of definite traces of bullet material, especially antimony, around the edges of a damaged area in bone in cases where no bullet or fragments could be found to establish the cause of a bone fracture. The technique was applied to establish the cause of death in a crime committed more than 20 years prior to the analysis.

Krishnan[50] and Coleman[63] have reported neutron activation autoradiographic techniques for examining bullet holes in materials.

Analysis of Ballistic Residue by "Activable Tracers"

In order to improve on the reliability of residue tests as mentioned above, activation analyses were performed on residues produced in test firings with gunpowder that had been deliberately tagged with readily activable rare earths, such as dysprosium and europium, and were reported by Guinn[6] and by Thomas et al.[81,82] It was possible to detect the tracers even at proportions of less than 0.1% in the cartridge primer. These techniques also suggest the further refinement of "coding" particular sources of firearms ammunition, explosives that might be used in criminal bombings, or other residues of forensic interest. This technique is outlined below.

Detection of Soil Transferred from Scene of Crime

The activation analysis of soil specimens from the scene of a crime found on the possessions of the criminal suspect has been reported by Pro et al.[64] and a link established between soil residues on a vehicle and the site of an illegal operation 1000 miles distant.

Trace-Element Patterns for Characterization of Forensic Material

The feasibility of characterizing a family of similar materials as to their particular source or unique past history through trace elements or incidental impurities ought, in principle, to exist for a wide variety of natural or synthetic materials, including those which

are common evidence materials associated with crime. The possibility of comparing specimens from the scene of a crime and those from the person or possessions of a criminal, and to establish with a high degree of certainty that they came from the same source, is potentially of great importance in forensic science and promises to open up many new avenues of investigation.

In a paper that outlines the great potential of FAA to modern criminalistics, Kirk has stated[53] that it should be possible to detect characteristic traces of almost every element in materials, provided sufficiently sensitive methods of analysis are available. In reality, most materials of natural origin, such as biological tissues from living organisms or those from synthetic manufacture, do contain a series of detectable trace elements or trace impurities at concentrations from a few parts per million down to parts per billion or less. Even synthetic materials produced under strict quality control have been found by high-sensitivity analyses to contain such impurities in variable amounts.

Moreover, since minor constituents tend to be incorporated in materials at concentration levels that depend on the type and purity of soil or diet on which living species depend or on the type of container materials, raw materials, catalysts, etc., involved in a synthesizing process, it is inevitable that wide variations in trace-element concentrations will be found in materials of a similar type, or of similar macroscopic composition but which come from different individuals, different geographical sources, different factories, or different production batches. This type of characterization might be most suitable for materials that are inherently uniform at some stage—e.g., those produced in batches, such as a furnace load of molten metal or alloy, a melt of glass, or a vat of a pharmaceutical preparation.

Techniques for Activation Characterization of Materials

There are several different methods that may be used to differentiate materials through trace-element content:

1. Radiochemical. A number of radionuclides can be radiochemically isolated from test specimens and their concentrations compared with controls.

2. Physical radiation analysis. Comparison of gross radiation and decay characteristics can be made for samples and controls on the basis that these features would be unique to each particular radionuclide mixture in activated samples.

3. Spectrometric. Instrumental spectrometric analysis of radionuclide mixtures can be carried out and quantitative comparisons of intensities at different energies made graphically, instrumentally, or by computer reduction of complex spectra.

All three of these methods have been used in activation-analysis "individualization" studies of different materials, and each has certain advantages and limitations. Radiochemical techniques are laborious, time-consuming, and destructive of evidence material; however, less matrix interference results; physical radiation techniques are less involved but are more empirical and less discriminating and therefore should be used only when gross differences in the composition of materials are known to be involved; spectrometric methods are also nondestructive and direct, more specific and detailed, but are still susceptible to matrix interference.

Activation Comparisons of Human Hair

It was evident from the earlier toxicological analyses of hair and nails (see above) that a number of trace elements were detectable in human head, body, and pubic hair, and the data available for arsenic content[14] showed a considerable range of content among the hair of different persons, while the content of a given individual's hair was quite uniform.

The basic feasibility of hair comparison by neutron activation was first presented by Jervis[54] in 1956 and demonstrated by the pattern of trace contents of arsenic, copper, and sodium among the hairs of four persons. It was quite evident that each of the four hair specimens could be clearly distinguished from the others merely by comparing the concentrations of the three trace elements.

Additional hair analyses revealed the presence of other trace elements such as Zn, Mn, Br, Se, Hg, and Sb, whose concentrations could be determined instrumentally through gamma-ray spectrometry.[55] The contents of 10 elements in about 90 hair samples were reported, and the ranges observed suggested the possibility of activation hair comparison for individualization. Nondestructive activation analyses were used in the comparison of a single fragment of hair found in a murder victim's hand with head hair from a prime suspect and from 25 other persons of similar age.[1,56] The presentation and acceptance of trace-element ratios in this investigation as evidence in a Canadian court in 1958 represented the precedent-setting introduction of activation analysis in a high court.

A large-scale hair survey for trace-element content was undertaken subsequently by Perkons and Jervis.[20,21] Samples consisting of hair bundles plucked mainly from the scalp and with root ends evident were collected in Canada and the United States, with a few specimens included from eastern Europe and India. The samples were representative of persons in different geographical location, persons occupationally exposed to pollutants, persons of all ages and occupations, family groups, identical twins, and mothers and their newborn infants. It was considered of interest to include many of the factors that might cause samples of different people's hair to be different or, conversely, to be similar.

It was found in these activation-analysis hair studies that the procedure for cleaning hair was quite important—since hair is usually contaminated with oily films, associated extraneous material, and sweat residues and since it was found that extensive washing removed appreciable amounts of some trace constituents from the hair and at the same time introduced increased amounts of others.[20] The effect of various cleaning procedures on the residual microconstituents of hair was investigated thoroughly by Bate and Dyer[57,58] using both natural hair and irradiated hair. They reported that an appreciable fraction of sodium and bromine, in particular, could be removed and should not be considered in activation hair comparisons for forensic purposes. The extent of variation among single hair strands from the same individual has been extensively investigated by Erickson, Krishnan, and Perkons[59]; the significance of the hair-growth cycle has been considered by Kerr,[60] and the possible effect of trace-element distribution along and among hair strands on hair-matching attempts has been investigated by Lima, Shibata, and Atalla.[61]

A large-scale, nondestructive neutron-activation-analysis study of the more than 1500 hair samples involved in the Perkons and Jervis survey[20] was carried out using sodium iodide detectors. Improved resolution in hair analyses can be obtained by using the Ge(Li) detector (Fig. 4). An approximate statistical analysis of the 30,000 hair analyses including quantitative data for 20 trace constituents indicated that it should be possible to use FAA to distinguish hair specimens, consisting of a few strands of several milligrams weight, with a high degree of certainty, provided more than 10 to 12 elements are determined. The estimated combined improbability of observing the same set of trace-element concentrations is given in Table 2.

A similar statistical study of trace-element content in hair from persons residing in England and Wales has been reported by Coleman,[62,63] and the results obtained from about 700 samples have tended to confirm the above conclusions. The analysis of 90 hair samples for trace elements was reported by Bate and Dyer,[57] and considerable differences were found among those from widely separated sources. The trace-element content of hair from ungulate and nonungulate animals has also been reported.

Evidence based on the neutron activation analysis of human hair strands has been presented to courts and

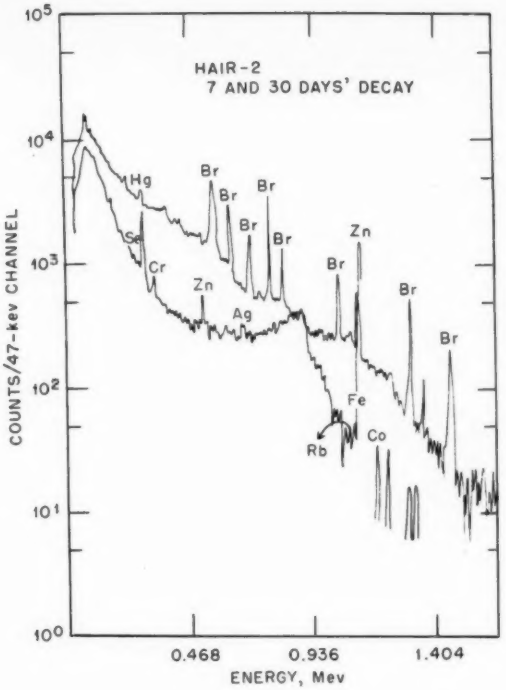

Fig. 4 Ge(Li) gamma spectrogram of neutron-activated human hair.

Table 2 Occurrence of Hair Trace Elements Throughout the Population

No. of elements analyzed (by order of ease of detection)	Improbability of chance occurrence of two similar hair microcompositions
1st 5 elements	26 in 100,000
1st 10 elements	3 in 10^7
1st 12 elements	5 in 10^9

Excluding sodium and bromine. (Computed from factors listed in Table 6, Ref. 20.)

has been accepted in Canada[1,86] and the United States.[64] Hair analyses by FAA have also been rejected on two occasions by U. S. courts in instances where only qualitative gamma-spectrometric comparisons of lightly irradiated hair were performed.

The international police organization, INTERPOL, solicited appraisals from several national police science bodies of the present status of activation analysis of hair and other evidence materials for presentation and acceptance in courts of law. Their findings were issued to member states as an INTERPOL document.[65] This document also presented a survey of qualified laboratories and research programs in the field.

Analysis of Fibers

While similar techniques should be applicable to natural fibers, little work seems to have been done. However, synthetic fibers used in textiles have been analyzed by neutron activation, and it was reported that the impurity contents of the fibers varied appreciably with the process and industrial equipment used for their manufacture.[83] The analysis of fiber fragments from apparel or from packaging and binding materials that are found in the investigation of crime should permit identification of the source of the fibers.

Activation Analysis of Tissues

In an analogous manner it should be possible to attempt matching of other tissues on the basis of trace-element patterns. Skin and nails are excretory tissues similar to hair in function, and some research has been reported[66] on the trace analysis of these tissues. Further studies with other tissues have been undertaken to assess the feasibility of relating dismembered parts of a cadaver, or tissue material derived from a wound.

Jervis and Wong[67] report activation-analysis procedures for trace analysis of more than 12 minor constituents of human blood (Fig. 5). The group-separation scheme uses 3 small ion-exchange columns and about 10 eluting solutions. The separations are performed without added carriers and are quantitative for most elements.

Even the use of the superior Ge(Li) gamma-ray spectrometer for the nondestructive analysis of blood (Fig. 6) does not diminish the necessity of using such radiochemical separation procedures to determine 10 to 15 of the more prominent trace elements in blood. These procedures are being applied to a trace-element survey of blood from different persons to assess the feasibility of matching blood found at the scene of a crime with bloodstains on the clothing or possessions of a criminal. The observation that the concentrations of some trace elements in blood vary rather rapidly in the same individual should not vitiate such comparisons of blood deposited at a given instant.

Characterization of Narcotic Drugs

Trace analysis by neutron activation analysis of 22 opiums from various illicit world sources was reported by Jervis et al.[1] and of 9 UN samples by Leddicotte[68] and by Bate et al.[69,74] Preliminary studies on the feasibility of characterizing other narcotic drugs as to origin were reported by Schlesinger et al.[70] A large-scale neutron activation analysis survey of trace impurities in seized heroins has been undertaken by Jervis and Rayudu since preliminary activation-matching comparison tests[71,72,80] indicated that 7 to 10 trace elements were present in illicit specimens in concentrations that varied considerably with source. The uniformity in impurity concentrations was quite good (ca. 10% variation) among groups of heroin capsules seized together. It was shown that seized heroins could be grouped quite distinctly according to trace-element-concentration ratios, and heroin from known foreign sources could also be classified within distinct groups.[73] Concentration ratios were used rather than absolute concentrations in comparing heroin samples because the extent of adulteration of heroin was variable among seizures and the water content was variable owing to the deliquescence of the material.

Similar trace characterization of a synthetic drug, Demerol, was also reported in the investigation of a sizable theft of the drug.[74]

Activation Matching of Physical Evidence—Glasses, Bullets, Paints, Soils, Liquor, Etc.

The activation analysis of glass fragments from automobiles such as are encountered in hit-and-run crimes was analyzed by Jervis,[75] and it was shown that replicate samples could be reliably identified with controls from gamma-spectrometric data. Good uniformity is found in the impurity content of batches of glass as reported by Coleman[76] and by Settle,[84] even over a period of time, in glass from one factory, but appreciable differences were observed among different sources of glass.[75]

Gamma-spectrometric analysis of neutron-activated bullet specimens was reported by Lukens and Guinn,[85,88] Different proportions of Sb, Sn, Cu, Ag, and Al were found in bullet leads from different

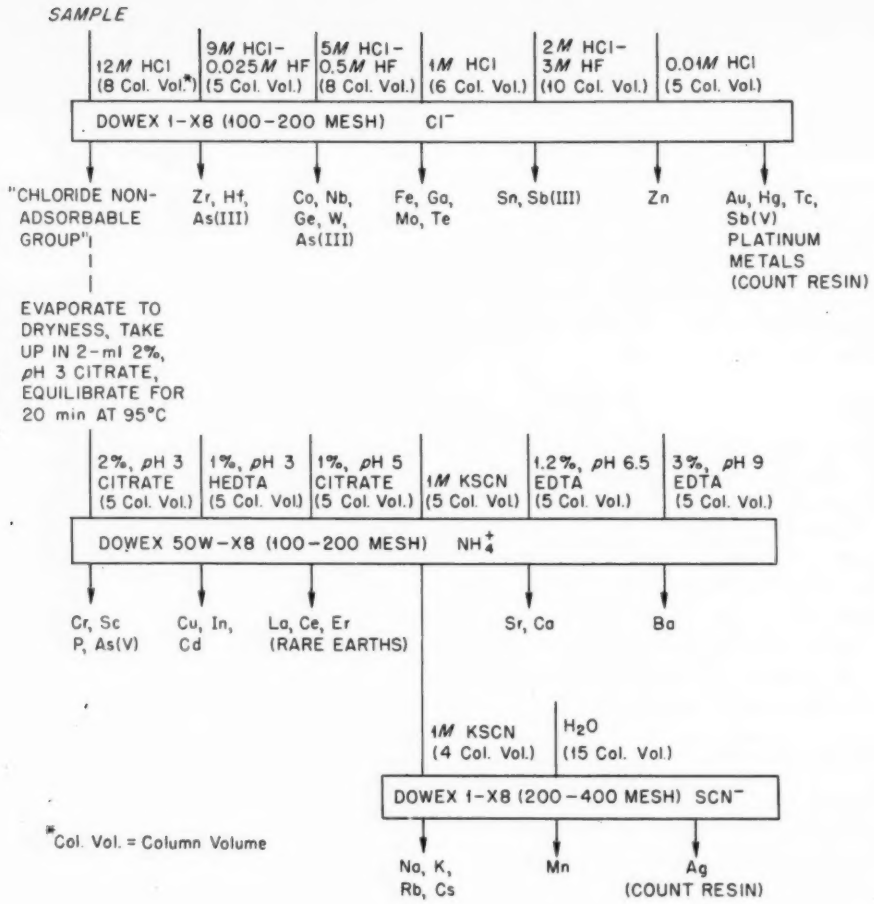
SAMPLE
12M HCl (8 Col. Vol.*)
9M HCl–0.025M HF (5 Col. Vol.)
5M HCl–0.5M HF (8 Col. Vol.)
1M HCl (6 Col. Vol.)
2M HCl–3M HF (10 Col. Vol.)
0.01M HCl (5 Col. Vol.)
DOWEX 1-X8 (100–200 MESH) Cl⁻
"CHLORIDE NON-ADSORBABLE GROUP"
Zr, Hf, As(III)
Co, Nb, Ge, W, As(III)
Fe, Ga, Mo, Te
Sn, Sb(III)
Zn
Au, Hg, Tc, Sb(V) PLATINUM METALS (COUNT RESIN)
EVAPORATE TO DRYNESS, TAKE UP IN 2-ml 2%, pH 3 CITRATE, EQUILIBRATE FOR 20 min AT 95°C
2%, pH 3 CITRATE (5 Col. Vol.)
1%, pH 3 HEDTA (5 Col. Vol)
1%, pH 5 CITRATE (5 Col. Vol.)
1M KSCN (5 Col. Vol)
1.2%, pH 6.5 EDTA (5 Col. Vol.)
3%, pH 9 EDTA (5 Col. Vol.)
DOWEX 50W-X8 (100–200 MESH) NH₄⁺
Cr, Sc P, As(V)
Cu, In, Cd
La, Ce, Er (RARE EARTHS)
Sr, Ca
Ba
1M KSCN (4 Col. Vol.)
H₂O (15 Col. Vol.)
DOWEX 1-X8 (200–400 MESH) SCN⁻
Na, K, Rb, Cs
Mn
Ag (COUNT RESIN)
*Col. Vol. = Column Volume

Fig. 5 Radiochemical group-separation scheme.

sources. Spectrographic analyses yielded similar results, and the two methods were considered complementary for use in bullet-matching attempts involving fragments or distorted bullets not amenable to normal ballistics tests.

Bryan, Guinn, and Settle[47,77] have reported the analyses of different paints with respect to the uniformity within batches and among manufacturers of similar paints. In the investigation of a burglary, a paint chip weighing less than 100 μg was removed from a metal tool and analyzed nondestructively by gamma spectrometry and compared with paint samples removed from a window.

Pro and Schlesinger reported matching of illicit liquors by activation analysis in conjunction with other means of characterizing the spirits.[64] Similarly they have compared soil samples found on a vehicle with specimens originating at the site of an illegal distilling operation 1000 miles distant and found similarities judged to establish that they had come from the same source.

A similar matching of plastic adhesive tape, of postage-stamp inks, of papers, and of typewriter inks has been reported from the same laboratory.

Incorporation of Activable Tracer Elements

To date most of the above characterization studies have been based on the incidental trace constituents incorporated in the materials of interest as a consequence of their particular origin. It has been suggested by Guinn,[78] Walgren,[79] and others that, at least in the

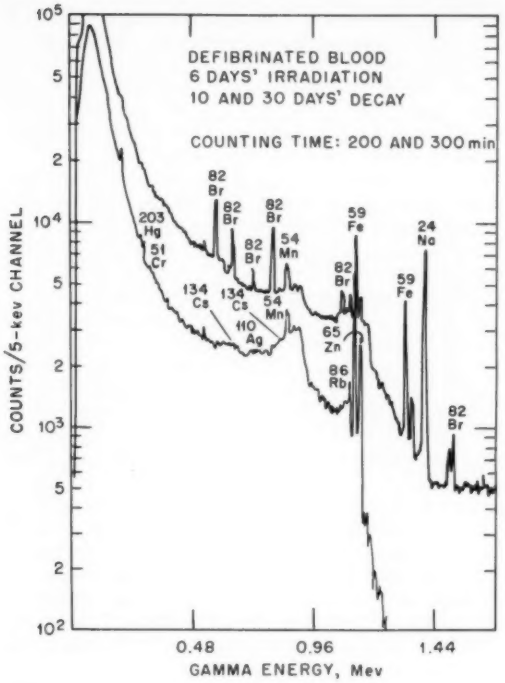

Fig. 6 Ge(Li) gamma spectrogram of neutron-activated defibrinated human blood.

case of synthetic materials such as pharmaceuticals and drugs, characterization can be facilitated by the addition of known concentrations (up to ~1 ppm) of several readily activable elements such as rare-earth elements that are not commonly found as impurities. This technique, if undertaken by legitimate manufacturers of materials subsequently distributed illegally, could greatly improve the feasibility of activation-analysis comparisons of such materials, derived from a criminal action, with reference specimens from known sources. Reference was made above to the tagging of firearms ammunition to facilitate the detection of gunshot residues and the identification of type and manufacturer of the ammunition.[81,82]

Conclusions

The foregoing review of published work on activation-analysis applications to criminalistics reveals the following definite achievements:

1. The determination of submicrogram quantities of phosphorus compounds and As, Hg, Se, and Tl in specimens from postmortem examinations and from living persons showing symptoms of toxicity has revealed injection of an abnormal amount of toxic substances by comparison with similar specimens from healthy persons. In some cases, with tissues such as hair and nails, the time scale of the ingestion of arsenic or mercury has been revealed through the distribution of the deposited element with distance from the growing end or edge.

2. Wounds, holes in apparel, and deposits on suspects' hands have been identified as arising from gunshots through the detection of increased traces of barium and antimony on bone, skin, or adjacent surface areas, and similar techniques have been applied to estimate the distance from a weapon to the target in cases of reputed suicide or homicide.

3. A series of feasibility studies on the possibility of distinguishing similar materials through their characteristic trace-element patterns has resulted from observations of the wide range or variation in trace-impurity content in specimens that come from different individuals or different natural sources. For example, extensive activation analyses for more than 20 elements in human head hair from many people have been carried out, and a statistical analysis of the results indicates that activation hair comparisons in forensic investigations may be quite definitive, provided sufficient hair sample is available and enough trace constituents are determined. Similar analyses of heroin, opium, marijuana, synthetic drugs, glass, paints, soil, and fibers have been made in attempts to characterize such materials according to source of supply. Methods have been developed for multielement analyses of blood in order to assess its individuality from person to person.

The incorporation of suitable marker elements in coded proportions allows the possibility of extending activation-analysis characterization of forensic materials.

The availability of high-resolution Ge(Li) solid-state gamma-spectrometry detectors has also extended the capability of nondestructive activation-analysis comparisons of activated specimens.

Activation analysis has the potential to contribute appreciably to the specialized and unique analytical requirements of criminology. Already a number of applications of the method have been made and techniques developed to the point that they may be applied in actual criminal investigations and presented as evidence in court. Further research on other promising applications is being undertaken, and it would appear inevitable that many forensic laboratories will undertake activation analyses in the future.

Acknowledgment

The author would like to acknowledge the contributions of colleagues who have assisted in forensic research and, especially, the cooperation of colleagues at the Ontario Centre of Forensic Sciences, Toronto, some of whose unpublished researches have been cited; the financial support of some forensic research by the Centre; and the heroin analyses by the Department of National Health and Welfare. (PSB)

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Note: GA-8171 in Refs. 8, 24, 28, 29, 51, 64, 76, 78, 79, and 84 refers to a report by Gulf General Atomic in San Diego.

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Tracing with Activable Tracers: Part II*

By Ralph E. Greene

Hydrology

The basic and theoretical aspects of the use of the stable-tracer activation-analysis method for water-resources investigations have been included in several reports[31-34] prepared under the sponsorship of the Division of Isotopes Development, U. S. Atomic Energy Commission. The selection of suitable materials and conditions—i.e., activable tracers, tracer nuclear properties, and tracer concentrations; procedures for sampling, activation, chemical separation, and measure-

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ment; corrections for background; sensitivity; and cost—is discussed in some detail, particularly in Refs. 33 and 34. The use of stable tracers and activation analysis is also discussed briefly in the International Atomic Energy Agency's *Isotope Techniques for Hydrology* (Ref. 35).

Open-Stream Flow Rates

Gardner and Dunn[36] demonstrated a technique for determining volumetric flow rates in open streams by introducing an inert tracer in the stream, taking samples downstream from the injection point, and determining the concentration of the tracer in the samples by activation analysis. The method did not involve introduction of any radioactive material into the stream and yet retained most of the high sensitivity and selectivity of radiotracer techniques. They stressed the fact that choice of the stable tracer is very important and that the substance chosen must satisfy the following requirements:

1. It must be nontoxic.
2. It should have a low natural concentration in the system under study so that the amount of added tracer required for detection can be reasonable.
3. It should be inexpensive and readily available.
4. It should be easily determined by activation analysis.
5. It should be free from interference of other materials in the stream and not be precipitated or adsorbed.

The stream on which they demonstrated their technique contained only two constituents easily activated by thermal neutrons: manganese and sodium. Average concentrations of these elements in the stream were 0.5 and 60 μg/ml, respectively. Since these elements are readily available in commonly occurring compounds and meet the other tracer criterions almost perfectly, they were evaluated as inert tracers. Weighed amounts of sodium chloride, sodium nitrate, and manganous nitrate were dissolved in water and introduced into the stream. One-milliliter samples, taken at 5-min intervals approximately 0.5 mile downstream from the point of tracer injection, were combined to give a composite sample for the test. Each single test required three samples for analysis by activation: (1) a blank for determination of the naturally occurring sodium or manganese, (2) a portion of the composite for determination of the average tracer concentration during the sampling time, and (3) a standard containing a known amount of tracer for activation comparison purposes. One-milliliter portions were irradiated 16 hr at a thermal-neutron flux of 5×10^{11} n/(cm^2)(sec) and 0.5-ml aliquots were placed 10 cm from a 2- by 2-in. NaI crystal and counted, using a multichannel analyzer. The ^{24}Na and ^{56}Mn gamma spectrums, corrected for decay, were analyzed by a least-squares matrix technique.

The results from the inert-tracer method were checked using the total-count radiotracer method[37] with ^{198}Au. Experimental results indicated that the activation method was valid under the existing test conditions; the maximum deviation from the radiotracer results was 6.4%, which is almost within the reported 5% precision of the radiotracer technique. Although the activation technique has some disadvantages (delay in obtaining results, requirement of rather large amounts of tracer, and uncertainty in the time of passage of tracer by the sampling point), the method should prove useful where the radiotracer technique cannot be used, and, of course, certain steps can be taken to minimize the disadvantages. To ensure that sufficient sampling time has been allowed, composite samples instead of one sample may be taken in sequence and compared with the blank to determine the last sample to be used for finding the average concentration. Also, to achieve higher sensitivity, the amount of tracer can be increased, larger samples can be taken, the samples can be irradiated in a higher flux, and the counting time can be increased. Chemical separations can also be used to improve sensitivity.

Kobayashi and various coworkers[38-43] have used a double-tracer technique for determining flow rate and dilution rate and, in some cases, for determining the velocity of rivers as well as for evaluating the nonactive tracer technique. In their work, a minimum amount of ^{24}Na was used as a radiotracer. Bromine was found to be a suitable tracer for the activation method, and natural bromine as NaBr or NH_4Br was used as a nonactive tracer. Bromine-82 as a radiotracer was preferred over ^{24}Na, ^{56}Mn, and ^{131}I since the adsorption loss was less for ^{82}Br (Ref. 43); however ^{24}Na was used as the radiotracer because the longer half-life of ^{82}Br—36 hr compared to 15 hr for ^{24}Na—was not suitable for the rivers under test since they were sources of potable water. (In these particular tests ^{82}Br would also have interfered with the activated stable bromide tracer.) In the experiments the radiotracer ^{24}Na was injected, as a pulse, into the stream, and immediately afterwards the nonactive bromide tracer solution was injected continuously for a given time at a constant flow rate. At various stations downstream, watertight scintillation counters signaled the arrival of the radioactive (and hence also the

inactive) tracer. During passage of the radiotracer, as indicated by the detector, samples were taken at regular intervals and were returned to the laboratory promptly to measure the ^{24}Na activity, by counting, for use in determining flow rate by the total-count method.[37] Likewise, during passage of the stable tracer, samples were taken at intervals for subsequent activation analysis and measurement of the stream flow rate by the dilution method.[44] In some of the experiments, a second quantity of radiotracer was injected into the stream to indicate the end of the continuous injection of the stable tracer. Fractions of the nonactive tracer-sample solutions were irradiated in a thermal-neutron flux tc form ^{82}Br by the $^{81}Br(n,\gamma)^{82}Br$ reaction. After irradiation the samples were allowed to cool for a number of hours before determining the ^{82}Br by multichannel gamma-ray spectrometry. In one variation of the method, 10 mg of NH_4Br was added to each sample after irradiation to supply carrier for the ^{82}Br in its precipitation as silver bromide. The precipitate from each fraction was counted in a multichannel gamma-ray spectrometer to determine the bromine tracer concentration.

The results of the test indicated that the stable tracer–activation method can be used to advantage in measuring velocity, flow rate, and dilution rate. Sampling is a very simple procedure. Dilutions down to 10^{-8} could be determined, compared with 10^{-6} for the radiotracer in amounts permissible in the river under test. The stable tracer–activation method proved to be more accurate under these particular experimental conditions; there was little interference due to activities produced from impurities in the rivers, which were fairly clean. Further sensitivity in the stable tracer–activation technique could be achieved by irradiating at a higher flux and using a larger quantity of stable tracer; however, unlimited quantities of even a stable tracer cannot be added to a stream used for drinking water.

Nagatsuka et al.[45] have also used bromine with the stable tracer–activation technique to study river flow, but with one significant modification—the stable bromine tracer was activated with 14-Mev neutrons rather than with thermal neutrons. The reactions of normal bromine with 14-Mev neutrons are

$$^{79}Br(n,2n)^{78}Br$$

$$^{81}Br(n,2n)^{80}Br$$

By counting the 0.51-Mev photons from annihilation of ^{78}Br positrons, the bromine content from both NH_4Br and $MnBr_2$ tracers introduced into the river water was determined. Manganese bromide was used to compare sorption losses of manganese and bromine. Interferences of the positron-annihilation photoelectric peak at 0.51 Mev were observed. The authors found the same interferences in deionized water samples as in river-water samples and observed that the decay period of the 0.51-Mev photopeak was in good agreement with that of ^{13}N. It was first supposed that the source of ^{13}N was the $^{14}N(n,2n)^{13}N$ reaction; however, boiling the water samples to remove dissolved nitrogen gas decreased the ^{13}N activity only slightly. They therefore assumed that most of the ^{13}N was produced by reaction of the recoil protons* with oxygen atoms in the water: $^{16}O(p,\alpha)^{13}N$.

Two procedures were used to determine the bromine—the first was to measure the activity of AgBr separated by precipitation and the second, a calculation method, was to mathematically resolve the decay curves of ^{78}Br (6.5 min) and ^{13}N (10 min). Samples were counted 3 and 8 min after irradiation, this delay being allowed for decay of ^{16}N (7.1 sec) formed by the $^{16}O(n,p)^{16}N$ reaction. River-water samples containing the NH_4Br were analyzed using chemical separation, and those containing $MnBr_2$ were analyzed using the calculation method, and the results of both methods were compared with those of the thermal-neutron activation analysis method. Tracer concentration values by the 14-Mev-neutron method were in good agreement with those determined by thermal neutrons. However, the standard deviation of the values from the fast-neutron activation method is greater than that in the thermal-neutron activation method, especially where the calculation method was used. This is possibly due to the activity from the ^{13}N being much greater than that from the ^{78}Br. Nevertheless, the use of 14-Mev neutrons does permit the determination of bromine in quantities greater than 1 ppm in river water and, in these experiments, the determination of dilutions of between $\sim10^{-5}$ to $\sim10^{-6}$ of the activable tracer injected into the river.

Leaks Under Dams

Hirayama and Osada[46] described leak tests on a dam that was considered to have widely branched underground outlets. These tests involved a double-tracer technique in which the radioactive $Na^{131}I$ and the activable tracer NaCl were used. The $Na^{131}I$ was added as a pulse to determine dilution by the total-count method, and the salt was introduced inter-

*Produced from the bombardment with 14-Mev neutrons.

mittently for determination of sodium concentration by activation. The flow rates of the leaks were estimated from these determinations.

Flows Through Porous Mediums

A patent,[47] entitled "Method of Tracing the Flow of Liquids by Use of Post-Radioactivation of Tracer Substance," has been awarded to F. E. Armstrong. The invention relates to a method for determining the flow of fluids through porous mediums, e.g., earth layers, particularly through oil sands of petroleum reservoirs. The fluid path through the porous medium is traced by first injecting a liquid containing a soluble salt of an element that is normally not present in the underground brine or water and then removing the fluid from another borehole at some distance from the first, irradiating the effluent fluid with neutrons, and measuring the activity produced in the added element (Fig. 1). The inventor suggests that Dy, Ir, Eu, and Sc are satisfactory activable elements but that other elements may also be used where desirable. Either an isotopic neutron source, e.g., Ra–Be, or an accelerator neutron source may be used. The latter produces a higher flux density than the usual Ra–Be source and offers no radiation hazard when not in operation.

The flow rate of liquid into and out of the irradiation vessel is controlled to maintain a constant rate of irradiation of the tracer since the amount of induced radioactivity is a function of both the amount of tracer present and the irradiation time. If no interfering activities are present, the activated tracer in the solution may be determined with a G–M counter, preferably a rate-meter type with a recorder. Should interfering activities be present, a scintillation counter with a pulse-height discriminator is suggested for measuring only one level of energy of radiation, i.e., that from the activated tracer. Should interferences at the same radiation energy levels as those of the activated tracer be present, the inventor suggests that simple separation may be made—either removal of tracer or removal of interfering substances from the liquid.

The patent states that a gaseous tracer may be used where the driving fluid is a gas; a finely divided solid tracer that can be dispersed in a gas or liquid medium may also be used.

Groundwater Characterization

Landström and Wenner[48] studied the possibilities of characterizing different groundwater supplies by

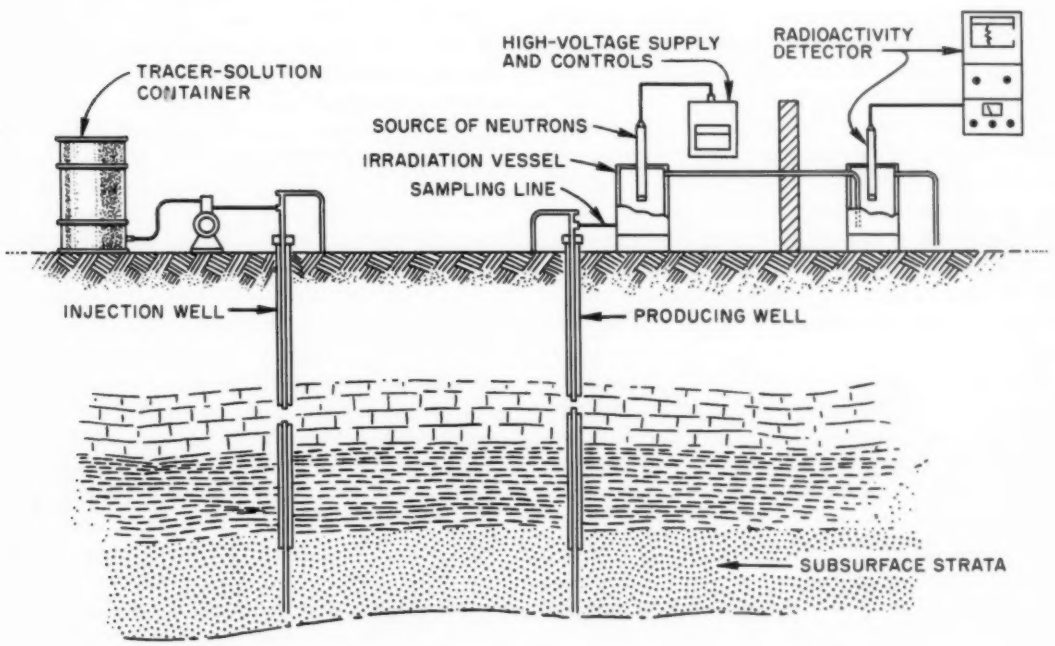

Fig. 1 Tracing fluid flow through a porous medium with activable tracers.

determining the natural trace-element content. They used a neutron activation method with chemical-group separation techniques. In the study of groundwater streams, they applied their method to the analysis of water from three locations in northern Sweden which illustrate common and important problems in hydrology. They studied different water supplies near the city of Skellefteå, and they also obtained information about the changes of water in water mains; in the valley of the Ume River they investigated the infiltration of river water and the location of the interface between the river water and the groundwater; and, at Sollefteå, they determined the presence and origins of different groundwater flows in a large excavation for a hydroelectric station. The authors concluded from the results that under certain geological circumstances there is no doubt that activation analysis of trace elements may characterize different groundwater supplies as well as reveal the presence of underground streams.

Photoneutron Method for Tracing Deuterium

A very interesting application of activable tracers for use in hydrology employs deuterium, a stable isotope of hydrogen, which is determined by counting neutrons produced by the photodisintegration of deuterium in the reaction $^{2}H(\gamma,n)^{1}H$. The neutrons produced by the reaction are counted, their number being proportional to the amount of deuterium present. The method, which appears to be ideal for water tracing since it uses an isotope of hydrogen that is stable, quite abundant, and inexpensive, has found limited application in hydrological studies because of the high threshold energy of the reaction—2.23 Mev. This high threshold combined with the requirement of a usefully long half-life severely limits the choice of a radioisotope as a source of photons for application of the method.

Haskell and Hawkins[49] used ^{24}Na as the high-energy (2.754 Mev) gamma source for tracing deuterium-tagged water in soil. Their ^{24}Na gamma source was contained, along with a slow-neutron detector, in a modified Nuclear-Chicago Model P-19 neutron-type depth-moisture probe. The outer metal shell containing the Ra–Be neutron source was replaced with a polystyrene shell containing 232 g of solid reagent-grade NaOH in a hollow cylinder. The shell was attached to the detector preamplifier with an adapter that permitted rapid attachment and removal. The NaOH was activated [$^{23}Na(n,\gamma)^{24}Na$] by attaching a threaded rod into the base of the polystyrene shell and inserting it in an irradiation port of a nuclear reactor at the Savannah River Laboratory. When removed from the reactor, the NaOH contained approximately 114 mc of ^{24}Na (15-hr half-life). With this amount of activity, it was possible to obtain accurate data throughout a 3-day period—9 mc of activity remained in the afternoon of the third day. The NaOH source could be reactivated as desired without building up long-lived sources of activity or inducing activity into the polystyrene shell.

When in operation the probe is lowered into an access tube in the soil, and the high-energy gamma rays from the ^{24}Na react with the deuterium in D_2O injected previously in the soil as a moisture tracer. The stable deuterium, upon bombardment by gamma rays, emits photoneutrons that undergo scattering and energy loss in the soil, and some of these slowed photoneutrons return to the detector and are recorded as counts on the attached scaler. The neutron count rate is proportional to the amount and distribution of the deuterium within the gamma field. A cylindrical lead shield housed the probe when above ground and could be positioned directly over the access tube in the field before lowering the probe. Personnel radiation exposure remained below 10 mr per 8-hr day.

The authors describe both a preliminary test and a field test as well as their method of calibration. Background counts were from two sources: (1) the gamma radiation from the source itself, which averaged 0.08 count/min per millicurie of source activity and (2) the natural occurrence of deuterium (0.0148 mole %) in water which varied in proportion to soil moisture content. Total background varied between 0.4 and 0.6 count/min per millicurie of source activity, while tracer peaks yielded up to 30 counts/min per millicurie. Measurements above 6.5 counts/min per millicurie had a statistical error of less than 5% at the 90% confidence level. They conclude that the method is both reliable and practical for observing the mass movement and semiquantitative vertical distribution of D_2O tracer in soil. To the degree that D_2O is a satisfactory tracer for water, the method provides a new and useful technique applicable to both laboratory and field studies of soil moisture. Placing tracers at several different depths about an access tube allows the study of soil-moisture movement over considerable vertical distance at a single site, and the use of a horizontal access tube would permit the study of lateral soil-moisture movement. Further developments are required before the method can be considered quantitative for rates of soil-moisture movement.

Improvements in the Photoneutron Method. The effectiveness of the photoneutron method for hydrological applications could be increased by using a longer-lived gamma source and higher source intensity. Although higher intensity sources would present shielding problems for field use, Wahl, Nass, and Kramer,* using multicurie ^{24}Na sources in hot-cell facilities, have been able to detect 50-μg quantities of deuterium. Their conservative calculations indicate that the lower limit of detection should be 0.5 μg with a 15-curie ^{24}Na source. Thus, as when tritium is used as a tracer in hydrological applications, deuterium-tagged water samples could be brought into the laboratory for analysis when a source too large for field application is used. Interferences from contaminants are rather limited. Beryllium (100% ^{9}Be) interferes directly[50-53] since it has a 1.67-Mev threshold energy for the $^{9}\text{Be}(\gamma,n)^{8}\text{Be} \xrightarrow{3 \times 10^{-16}\ \text{sec}} 2\alpha$ reaction, which is lower than that for deuterium. Lithium, boron, chlorine, and cadmium interfere indirectly because these elements have high thermal-neutron cross sections and may capture some of the moderated photoneutrons before they reach the neutron detector.

Haskell and Hawkins[49] also indicated that an obvious improvement in the deuterium–photoneutron method would be the substitution of a high-energy, long-lived gamma source for ^{24}Na. They suggest the possibility of using ^{208}Tl (2.62-Mev γ, 3.10-min half-life) which is in equilibrium with its parent, ^{228}Th (1.91-year half-life).† Thorium-228 sources‡ are now commercially available; however, one must take into consideration both the source compound (if the source is not in the metallic form) and the impurities so as to reduce to a tolerable level the neutron background resulting from the reaction between elements of low atomic number and energetic alpha particles emitted by the ^{228}Th and its daughters.

In a further search for a suitable source, if one examines gamma-ray energies above the 2.23-Mev threshold level in tables[54,55] arranged according to increasing gamma-ray energies from various radionuclides and also considers half-life and percentage of disintegrations having gamma rays of sufficient energy, he finds that only one nuclide—^{56}Co—meets the requirement to a reasonable degree. Cobalt-56 has a half-life of 77.3 days, and above the D(γ,n) threshold level it has gamma rays of 2.60 Mev (16%), 2.99 Mev (1%), 3.25 Mev (12%), and 3.47 Mev (1%). This cyclotron-produced nuclide has been prepared at ORNL,[56] § but, although sufficient yield data are not available for a precise cost estimate, the price is not expected to be low.

One other possible source of high-energy photons has been described by Plaksin, Malyshvea, and Starchik,[57] who used the bremsstrahlung radiation from high-energy beta emitters to induce photonuclear reactions with beryllium and deuterium. They suggest the use of ^{106}Ru (367 days) in equilibrium with its ^{106}Rh daughter (30 sec) that has a beta emission of 3.54 Mev maximum energy. Another source material is ^{144}Ce (285 days) in equilibrium with its daughter ^{144}Pr (17.3 min) with a maximum beta energy of 2.99 Mev. Both of these beta emitters have some bremsstrahlung radiations energetic enough to initiate the photonuclear reaction with deuterium; however, as in the case with the beta spectrum itself, the quantity is small at the higher energies. These workers found that high contents of beryllium or deuterium can be determined with a relatively weak source of hard photons; unfortunately, when low contents of beryllium or deuterium are to be determined, source activities higher by a factor of several hundred or more are required. The yield of neutrons from a ^{106}Rh–^{106}Rh bremsstrahlung source is much, much less than from a ^{24}Na source of equivalent activity.

The development of ^{228}Th, ^{56}Co, and possibly the bremsstrahlung sources offers interesting possibilities for the determination of a hydrogen isotope—deuterium—in the field. Of the three longer lived sources, ^{228}Th (1.9 years) with its ^{208}Tl daughter appears to be the most promising. At the present stage of process development, ^{56}Co with its 77.3-day half-life is too expensive to be considered as a gamma source for hydrological applications for the photoneutron reaction with deuterium. It also appears that the low efficiency of the bremsstrahlung sources makes them unsuitable for this application.

(Part III of this series will appear in a subsequent issue.)

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Section

III

Isotopes and Radiation Technology

Food Irradiation

The Commercial Prospects for Selected Irradiated Foods

By D. M. Yates, G. W. Collings, I. L. Kinne, J. R. Kircher, J. H. Litchfield, and O. Wilhelmy, Jr.

Abstract: *A brief summary is presented of a recent report on the commercial prospects of selected irradiated foods, based on studies done at Battelle Memorial Institute. The outlook for commercialization, at the time of this study, for radiation pasteurization of marine products and radiation improvement of dehydrated vegetables is good. Radiation-treated beef, pork, and poultry need further investigation.*

In January 1965 the Business and Defense Services Administration of the Department of Commerce published *Current Status and Commercial Prospects for Radiation Preservation of Food.* It was intended to stimulate industry interest in radiation processing of food. The report* summarized here is an extension of that study; the work was done at Battelle Memorial Institute. The study's major objectives were to determine the commercial prospects for selected irradiated foods by analysis of relevant factors, to recommend action to expedite commercialization, and to present the results in a report designed for managers in food and related industries.

The foods and processes chosen for the study were: radiation-improved dehydrated vegetables; radiation-inhibited ripening of bananas; radiation-pasteurized seafood; radiation-pasteurized beef, pork, and poultry; and radiation-sterilized beef, pork, and poultry. Hypotheses were developed about the use of radiation, information was collected to test the hypotheses, and the information was analyzed. Factors favoring or limiting the application of radiation treatment in the food industry were identified.

*D. M. Yates, G. W. Collings, I. L. Kinne, J. R. Kircher, J. H. Litchfield, and O. Wilhelmy, Jr., The Commercial Prospects for Selected Irradiated Foods, USAEC Report TID-24058, March 1968.

Cost Factors

Variables that affect the cost of irradiating food are the product itself, the radiation dose required, the throughput, the source efficiency, and the ease of fitting the radiation process into the existing process scheme. In general, irregularities of product shape, density, and size reduce efficiency and increase costs. Throughput affects cost by dictating a source large enough to deliver a certain dose to a given number of items in a stated period of time.

One of the greatest costs of a radiation processing plant is the radiation source itself. The two radioisotopes most likely to be used are ^{60}Co and ^{137}Cs. Machine irradiators were also evaluated; they may have an advantage when high dose and high throughput are combined with a thin, uniform product configuration. Because of the lack of commercial operating data, operating costs were estimated from radiation processing experience in other applications and from engineering handbook procedures. Costs based on several different variables were derived. For example, a 125-Mrad-lb/hr plant, pasteurizing with a 40% efficiency and using ^{60}Co costing 35 cents/curie, was estimated to have a plant cost of $260 thousand, a source cost of $9 thousand, and a yearly operating cost of $131,600.

Individual product treatment costs were estimated also. These ranged from a low of 0.12 cent/lb for inhibiting ripening of bananas by radiation to a high of 15.3 cents/lb for sterilizing beef by radiation. The

possible need for cryogenic facilities would raise the estimate for sterilizing meat by about 2 cents/lb.

There are other economic factors affecting commercializing the radiation processing of foods. One of these is the cost of consumer education. Although a survey in late 1966 indicated that the majority of consumers would accept any process given Food and Drug Administration (FDA) approval, there is an active minority who might oppose marketing of irradiated food. Another factor is the reluctance on the part of investors created by lack of FDA approvals, the uncertainty about consumer response, and the newness of the process. Radiation facilities must compete with other uses for the risk capital available to the food industry, which is composed of many small firms with relatively little risk capital.

Radiation-Improved Dehydrated Vegetables

A patent was granted to Thomas J. Lipton, Inc., on Mar. 13, 1962, covering improvement of tenderness and rehydration characteristics of dehydrated vegetables by irradiation. Irradiating dehydrated vegetables decreases their rehydration time from the present 10 to 15 min to 2 to 4 min. Other benefits are increased tenderness and equalization of cooking times for mixed ingredients.

Inhibition of Ripening of Bananas

Research has shown that a dose of 25 to 50 krads delays ripening of bananas 8 to 26 days. Canadian researchers have shown that anthracnose* spoilage could be eliminated by a 300-krad dose. However, industry is investigating alternative methods of inhibiting ripening, such as control of storage atmosphere and introduction of various packaging innovations. The banana industry is fairly efficient at present, and claims paid on losses in distribution amount to only 0.375% of gross sales. The margin between benefits and costs is evidently small.

Sterilized and Pasteurized Poultry

A 4.5-Mrad dose will destroy virtually all microbes in poultry meat. A dose of 0.1 to 0.25 Mrad reduces the microbial load and gives a 2-week extension of shelf life at refrigeration temperatures. Radiation sterilization of poultry meat would mean the marketing of shelf-stable items that could be handled like any other dry grocery product. A further benefit would be the elimination of the ice-water-bath step in processing carcasses, which is a slow step because of the low heat-transfer rates involved. The principal benefit from radiation pasteurization of poultry meat would be greater inhibition of *Salmonella*. Even though this microorganism is killed by low heat, some microbes could be transferred from poultry to other, uncooked food during handling in a restaurant or home, e. g., by using the same knife to cut chicken and to slice bread.

Irradiated Beef and Pork

The research on radiation-sterilized beef and pork has been carried out mostly at the U. S. Army Natick Laboratories; this reflects the Army's interest in irradiation of food as a logistics improvement. The Army's goal is the development of stable products with a shelf life at room temperature of 1 to 3 years. This goal can be achieved. Some problems facing radiation sterilization of fresh red meat are its altered properties: moisture loss and change of color and physical appearance. Some interest in radiation pasteurization of fresh red meat has been shown by sellers who think the 2-week extension in shelf life would give them greater marketing freedom.

Pasteurized Marine Products

Research at the Marine Products Development Irradiator, Gloucester, Mass., has shown that a dose of 0.10 to 0.25 Mrad extends shelf life of many seafoods by 2 to 3 weeks at temperatures of 0 to 3°C. Test panels and troop feeding studies have shown that the irradiated fish is organoleptically acceptable. Radiation can also preserve products that would spoil before they could be processed. The benefits from radiation pasteurization of seafood would be fresh fish markets inland and easier purchasing by grocers (i.e., they would have greater variety, greater supplies, and less worry about underbuying or holding sacrifice sales). Industry has shown a definite interest in radiation pasteurization of fish. Market development would probably spread inland from areas where fresh fish are known and preferred. (FEM)

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Food-Irradiation Activities Throughout the World[1]

By J. H. Clausen* and J. W. Osburn, Jr.†

The food-irradiation activities of research organizations throughout the world have been summarized in a recent report of the Business and Defense Services Administration of the U. S. Department of Commerce. The publication has two objectives: (1) to identify the general dimensions and scope of the food-irradiation programs throughout the world and (2) to provide a reference that may facilitate further exchanges of technical knowledge and techniques among interested parties. The report is divided into three parts:

Index of Overseas Food-Irradiation Programs and Capabilities. In the first part the governing organization of each country is listed, and details are given on each laboratory where research is in progress or planned. The 76 countries listed account for almost 80% of the world's population. Of these, 50 have provided scientific and technical staff with equipment and facilities and have accomplished substantial progress in food-irradiation research. In the remaining 26 countries, organizational plans and structures are complete, and programs are being outlined. For each entry under a country, the name of the laboratory is given in boldface type, with the full address of the laboratory; listed under the laboratory are whom to contact, what the laboratory's function is, what equipment is available, and what programs are in progress.

Outline of Food-Irradiation Programs in the United States. The second part reviews the activities taking place in the United States and gives a guide to the reference materials and sources of information currently available. All research on irradiation of food in the United States is controlled by the government, based on three statutes: (1) The Federal Food, Drug, and Cosmetic Act, with amendments, which treats ionizing radiation as a food additive; (2) The Federal Meat Inspection Act; and (3) The Poultry Products Inspection Act of 1957. Under these regulations, particularly the first one, several foods and specific treatments have been approved. These are radiation sterilization of bacon (February 1963), radiation disinfestation of wheat and wheat flour (August 1963), radiation inhibition of sprouting of white potatoes (July 1964), and various packaging materials (August 1964 to July 1967).

International Organizations. In Part III, 20 international organizations are listed which have either programs or an active interest in food irradiation. Some of these, like the International Atomic Energy Agency in Vienna, have several programs going at their own

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facilities; others, such as the World Health Organization of the United Nations, do not have their own programs but support research by conducting international meetings on the subject of food irradiation.

This report is a valuable guide to the activities taking place all over the world in research into the possibility of using radiation to process food. If the efforts of these 77 countries and 20 international organizations are successful, the world food supply could perhaps keep pace with the world population growth. (FEM)

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Section
IV
Isotopes and Radiation Technology

Process Radiation Development

Radiation-Induced Solid-State Polymerization*

By J. H. O'Donnell†

Abstract: *An increasing number of monomers have been shown to polymerize in the solid state. Gamma irradiation is the most useful method of reaction initiation because of its high penetration and relatively uniform distribution of active sites. Polymerization may occur by free-radical, cationic, and possibly anionic mechanisms. Electron spin resonance is a useful tool for following the course of free-radical reactions in the solid. Many polymerizations occur at phase changes, which are conveniently detected by differential thermal analysis. Some solid-state polymerizations are hindered by the existence of a crystalline lattice, but others are assisted. There is as yet no satisfactory picture of the kinetics of the solid-state polymerization process.*

High-energy radiation, such as gamma rays, will initiate polymerization in a variety of solid monomers. Free-radical, cationic, and anionic polymerizations have all been suggested as occurring. Polymerization may take place during irradiation or on subsequent warming. Experimental difficulties arise in following solid-state reactions, and increasing use is being made of non-destructive techniques, such as electron spin resonance, differential thermal analysis, and X-ray diffraction. The role of the crystalline lattice of the monomer remains a subject of controversy; in some monomers it appears to assist the reaction but in others to hinder it. Vinyl monomers usually yield amorphous polymers, but some cyclic monomers have given partially crystalline polymers oriented relative to the monomer lattice. Investigations of radiation-induced solid-state polymerization are providing information on the mechanism of radiolysis and the mobility of molecules and radicals in organic solids. Industrial interest stems from the possibilities of producing polymers from new monomers and of improving the structural properties of existing polymers.

The manufacture of plastics, synthetic fibers, and synthetic rubbers has increased rapidly since the beginning of this century and is now one of the major industries of developed countries. Most of this advance has occurred in the 20 years since the end of World War II and shows no sign of slackening. The basic feature of these three fields is the preparation of polymer molecules, made up of long chains of monomer units.

A very recent trend is toward a greater diversity of monomers and the use of combinations of these in several ways in each polymer molecule. Also, an increasing degree of control can be exercised over the spatial arrangement, or steric structure, of the atoms in each molecule; such stereoregular polymers have enhanced physical properties, including higher softening temperatures and greater mechanical strength. To an increasing extent, polymers can be "tailor-made" to give the properties desired for any particular application. The demand for an increase in the number of possible monomers and for a variety of steric structures has led to an intensive search for new methods of polymerization. One of the most interesting of these is polymerization in the solid state—a process that has been widely studied[1] but whose nature is far from being completely understood. It is also a subject of considerable intrinsic interest in view of the attention now being given to the organic solid state.

High-energy radiation is the only widely applicable method of initiating solid-state polymerization and is virtually the only controllable way of polymerizing large quantities of monomer. The increasing availability of radiation sources, such as spent reactor fuel ele-

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ments,[2] ^{60}Co, and electron accelerators,[3] has led to widespread research into solid-state polymerization and will help to make industrial processes technically and economically feasible.

Initiation

Polymerization can be initiated in a number of ways in the solid state. Some monomers can be polymerized simply by heating. These include vinyl monomers such as acetamidostyrene,[4] benzamidostyrene,[5] and *p,p'*-divinyldiphenyl;[6] condensation polymerization of α,ω-amino acids,[7] ϵ-amino caproic acid,[8] and alkali metal *p*-halothiophenates[9] is also possible. In fact, most condensation polymers probably reach an equilibrium molecular-weight distribution in the solid state near the melting point.[10,11]

Chemical initiators are used in most liquid-phase polymerizations, but they are not satisfactory with solid monomers. Initiators that decompose thermally to produce free radicals, e.g., peroxides, would have to be stable at the melting point of the monomer, in which case the rate of initiation in the solid would be negligible. An initiator that decomposed at a reasonable rate in the solid monomer could not be added to the liquid monomer, prior to freezing, without immediately initiating polymerization. Similarly, it would be impossible to incorporate an ionic initiator into the solid without causing polymerization during the liquid-phase mixing. There are two ways of overcoming this difficulty. A thermally stable chemical initiator can be incorporated into the solid and decomposed by radiation,[12] or the polymerization can be initiated heterogeneously at the surface.[13]

Ultraviolet light will initiate free-radical polymerization in the liquid phase but has very limited use for solids. Absorption of the light occurs at the surface, and the reaction proceeds slowly inward; consequently this method can be used only on thin layers and has been restricted to some academic studies.[14]

High-energy irradiation, e.g., with gamma rays, is a very convenient method of initiation, which gives a relatively uniform distribution of active sites for initiation of polymerization throughout large quantities of monomer. It can be used for a wide variety of monomers, since cationic, anionic, and free-radical centers are all produced. Recent appreciation of the importance of electrons in the radiolysis of organic solids as well as aqueous solutions suggests that anionic polymerization may be important in suitable monomers. Initiation by radiation may be summarized as:

Cationic: $R \rightarrow R^+ + e^-$

Anionic: $R + e^- \rightarrow R^-$

Free radical: $R^+ + e^- \rightarrow R^* \rightarrow R^1\cdot + H\cdot$

$$\begin{array}{c|c} | & | \\ C{=}C & + H \rightarrow \end{array} \begin{array}{c|c} | & | \\ -C & -C\cdot \end{array}$$

Dose rates in the range 0.01 to 1 Mrad/hr, which are convenient for solid-state polymerization, can be obtained from spent reactor fuel elements and ^{60}Co sources. Higher dose rates (up to 10 Mrads/min) over small areas can be produced by electrons from electron accelerators and from X rays produced by impingement of the electron beam on a suitable target. The electrons penetrate only a few millimeters into solids and give very high absorbed doses, whereas X rays are less readily attenuated and therefore give lower absorbed doses per unit time.

Radiation initiation is the best method for producing controlled polymerization by free-radical and ionic reactions in large quantities of solid monomer; hence it is likely to be the basis of future industrial processes.

Polymerization

There are two methods of polymerization in the solid state using radiation initiation.

The first is "in-source" polymerization, which consists of irradiation at a temperature at which polymerization takes place at a sufficient rate during irradiation. The conversion to polymer follows a sigmoid curve with time, i.e., there is an induction period, followed by a very rapid increase in polymerization rate and finally a tailing off. This is illustrated in Fig. 1, which shows conversion curves for the polymerization of methacrylic acid (m.p. 15°C) at 0°C during irradiation with gamma rays at different dose rates.[15] Very high conversions are frequently obtained during in-source polymerization, but practical difficulties arise when appreciable quantities of monomer are used, owing to heat buildup during the period of rapid polymerization.

The effect of temperature is extremely variable. A very strong temperature dependence ($E_a \sim 40$ kcal/mole) has been observed for trioxane,[16] a lower value for styrene[17] ($E_a \sim 9$ kcal/mole), even lower values for acrylonitrile[18] ($E_a \sim 3$ kcal/mole), and a negative value for cetyl methacrylate[19] ($E_a \sim -0.7$ kcal/mole). In many cases low polymerization rates have been reported down to very low temperatures, but, since

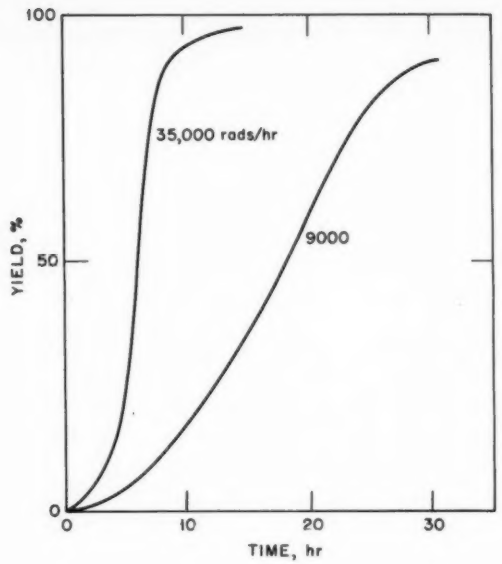


Fig. 1 In-source polymerization of methacrylic acid in the solid state by gamma irradiation at 0°C.

only small yields of polymer are obtained, the reliability of these results is frequently questionable. Nevertheless, it seems possible that some polymerization may occur as low as −196°C, probably as a result of the absorption of radiation energy to cause molecular excitation. These results have led to ideas of a type of "electronic" polymerization.[20,21]

The second method is "postirradiation" polymerization, which is carried out by subjecting the monomer to a suitable total dose of radiation at a temperature where polymerization is negligible. The monomer is then warmed to a temperature at which polymerization takes place at an adequate rate and with a suitable degree of conversion. The rate of postirradiation polymerization at any temperature is rapid at first but then decreases to a low value and continues for an extended period. Typical curves[22] for polymerization of anhydrous zinc methacrylate after irradiation at −196°C are shown in Fig. 2. The conversion at which the polymerization rate tails off, i.e., the limiting conversion, depends on the particular monomer, the radiation dose, and the temperature.

When large samples are used, the initial rapid polymerization may generate excessive heat, particularly in strongly exothermic vinyl polymerizations. This heat cannot be conducted away through the monomer, which actually will be an efficient thermal insulator. Consequently the interior temperature rises, the rate of polymerization increases, and a classical thermal explosion results. Such explosions have been reported in acrylamide[23] and formaldehyde[24] but, in fact, are difficult to avoid with large quantities of any polymerizable monomer.

Postirradiation polymerization should be simpler to study than in-source polymerization because the initiation reaction has been separated from propagation and because termination has probably been eliminated. Solid-state polymerization is usually followed by separating the polymer from unreacted monomer after various periods of irradiation or postirradiation heating. The monomer is dissolved in a solvent that does not dissolve the polymer, or both monomer and polymer are dissolved in a solvent, and then the polymer is precipitated by addition of a nonsolvent.

This technique of differential solution has two serious disadvantages. First, the dissolution of the monomer takes a finite time, and the rate of polymerization is known to be greatly enhanced during any phase change. Thus it is possible for a significant amount of polymerization to take place at this stage. Careful measurements should be made to estimate the importance of this effect in every case; it is quite likely that many reported polymer yields from solid-state polymerization are seriously in error due to this effect. Second, it is difficult to obtain solvents that will

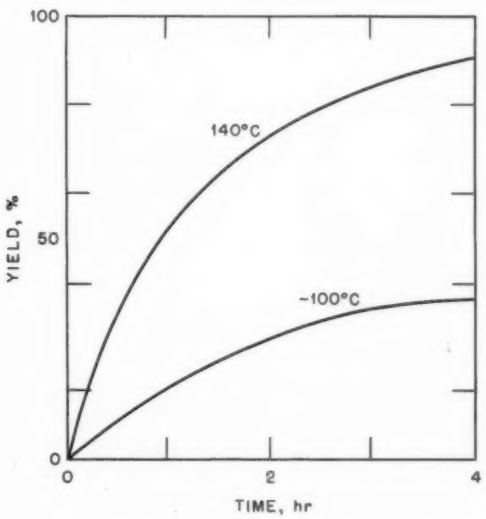


Fig. 2 Postirradiation polymerization of anhydrous zinc methacrylate in the solid state; 1 Mrad of gamma irradiation at −196°C.

distinguish between monomer and short-chain polymer, and there is evidence that separation by differential solution gives only the yield of long-chain polymer.[25]

Obviously a nondestructive method of measuring polymer content in situ is required. This has not been achieved yet. The loss of birefringence in wafer-like single crystals has been used as a measure of the loss of monomer crystallinity in acrylic and methacrylic acids.[14] The X-ray diffraction patterns of acrylamide during solid-state polymerization have indicated that the unreacted monomer retains its crystal structure and that the polymer is amorphous.[26] However, this method has not been used for quantitative estimation. Differential thermal analysis has been used to observe high rates of polymerization and the sudden onset of polymerization on warming.[27]

Topochemistry

Control of a chemical reaction in the solid state by the crystalline lattice is well known. This "topochemical" effect may influence either the rate of the reaction or the nature of the products. Such phenomenons[28] are well known in both inorganic and organic solids. For example, the radiation-induced decomposition of choline chloride,

$$[(CH_3)_3NCH_2CH_2OH]^+Cl^- \rightarrow [(CH_3)_3NH]^+Cl^- + CH_3CHO$$

is much faster in the orthorhombic solid below 73°C than in the cubic form above 73°C or in the liquid.[29] Again, when tetraglycine methyl ester is heated in solution, it loses methanol to form polyglycine, whereas in the solid state, sarcosyltriglycine is produced.[30] Undoubtedly, some of the interest in solid-state polymerization arises from the possibility that similar differences between liquid- and solid-phase reactions may be discovered.

Crystalline polymers have been obtained from isocyanates by radiation-induced polymerization in the solid, but not in the liquid, state.[31] Gamma irradiation of diketene in the liquid phase produces mainly β-lactone, but in the solid (at $-78°C$) a linear polyester results.[32] An unusual polymerization reaction, involving intermolecular bond switching, which depends uniquely on the spatial arrangement of the monomer molecules, has been reported.[33]

Different polymerization rates have been observed[34] in different isomorphs of tributylvinylphosphonium, $[CH_2=CHP(C_4H_9)_3]^+$, bromide.

There is some evidence also that the stereoregularity of the polymer may be affected, e.g., the atactic content is greater in poly(methacrylic acid) (Ref. 35) and poly(barium methacrylate) (Ref. 36) produced in the solid state than in the liquid at low temperature.

The question of whether polymerization actually does occur in the solid state is pertinent since many monomers show only postirradiation polymerization at appreciable[35,37,38] rates in the solid state near their melting points. Local heating, due to the exothermic heat of reaction, could produce a microscopic packet of liquid at the growing end of the chain. This idea was first advanced for inorganic solid-state reactions[39] but is equally possible in organic solids.

In-source polymerization frequently occurs at lower temperatures, but in this case the energy absorbed from the radiation may assist in the local melting. The concept of "pseudoliquid" polymerization is supported by the fact that monomers irradiated at low temperatures often polymerize on warming at temperatures corresponding to phase changes in the solid when there is a high degree of disorder and molecular mobility. The importance of solid–solid phase changes was first noticed in inorganic solids,[40] but these changes are now being considered inorganic solid-state polymerization.

Another idea, which is very difficult to refute, is that polymerization occurs by sublimation of monomer molecules across the cavity formed at the end of the growing chain by contraction during bond formation, i.e., a "pseudogas" reaction.

There are some systems in which it is extremely unlikely that localized melting or vapor transport occurs. For a series of metal salts of acrylic and methacrylic acids, Morawetz and coworkers[36,41] found that polymerization took place in some cases, e.g., potassium acrylate, more than 200°C below the melting point but did not occur at all in some salts, e.g., lithium methacrylate. The polymerizability did not show the same correlation with the metal ions in the acrylates and methacrylates, indicating that the crystal structure of the monomer was the deciding factor. This was supported by the great variation in behavior of different hydrates of the same salt, which was quite unrelated to the amount of water present.

Also, some monomers, e.g., styrene[37] and methacrylonitrile,[42] show a greater rate of polymerization in the solid near the melting point than in the liquid phase just above the melting point. However, this may be due, at least in part, to suppression of the termination reaction in the solid.

There are other monomers for which the rigidity of the crystalline lattice is an obvious impediment to polymerization. Vinyl acetate polymerizes more rapidly as a glass than in the crystalline form.[43] Methyl methacrylate does not polymerize to any extent in the pure solid, but a frozen mixture with paraffin oil polymerizes readily.[44]

Mechanism

Polymerization reactions in irradiated solid monomers may occur by free-radical, cationic, or anionic mechanism, since all three types of active sites may be produced by irradiation. Unfortunately the methods of discrimination used in the liquid phase are of doubtful value. If a free-radical mechanism is suspected in a liquid-phase reaction, then a free-radical scavenger is added, and the polymerization should be inhibited or retarded. Such methods have been used in the solid state,[37,45] but either the scavenger will be expelled into pockets by the monomer crystals during solidification and will not be effective, or, if it is uniformly distributed, it will seriously alter the perfection of the lattice, and this may be sufficient in itself to disrupt the growing chains, irrespective of the mechanism. For example, any additive, independent of its chemical nature, has been shown to lower the polymerization rate of acetaldehyde.[31] One inhibitor of interest is oxygen, but its effectiveness will be governed by the permeability of the lattice as much as by chemical considerations. Retardation due to oxygen in the in-source solid-state polymerization of methacrylic acid is shown in Fig. 3. Oxygen also inhibits polymerization in zinc methacrylate[22] but not in barium methacrylate[36]; this has been attributed to a difference in permeability of the solids.

The addition of chemical initiators to test whether different types increase the rate of polymerization is precluded as discussed earlier.

Therefore it is usual to assume that the reaction proceeds by the same mechanism as in the liquid phase. Sometimes indirect evidence can be obtained, e.g., acrylic and methacrylic acids will polymerize in the solid state under ultraviolet illumination[14] or gamma irradiation. However, although the ultraviolet-initiated reaction is almost certainly a free-radical one, this does not prove that the gamma-initiated reaction is, also. Similarly, trioxane can be polymerized in the solid by contact with BF_3 vapor[46] or a solution of $SnCl_4$ in *n*-hexane,[47] indicating a cationic mechanism and suggesting that the radiation-induced polymerization is also cationic.

Likewise, the heterogeneous initiation of polymerization in α-phenyl-β-propiolactone by *n*-butyl lithium in ether[48] suggests that an anionic radiation-induced reaction is possible.

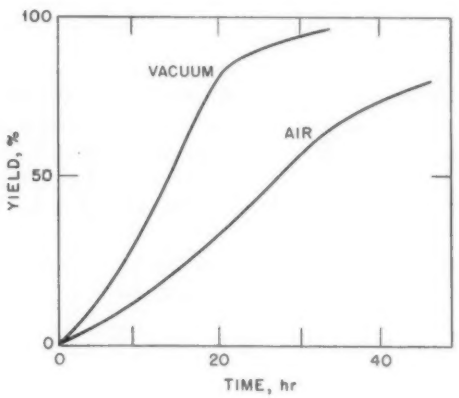


Fig. 3 Effect of oxygen on postirradiation polymerization of methacrylic acid.

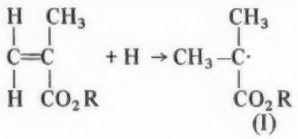
Zero or negative activation energies for polymerization in the solid are sometimes taken as evidence of an ionic mechanism.[49] This is reasonable for the liquid phase (it is due to a decrease in the rate of termination at low temperatures), but it is not convincing in the solid where the termination rate is almost certainly controlled by physical parameters. Furthermore, the very low conversions usually reported for polymerizations at low temperatures are suspect because of the tendency for polymerization to occur during melting or dissolution.

Electron Spin Resonance

A major difficulty in examining any chemical reaction in the solid state is to observe the system in situ. Until a few years ago, a similar situation prevailed in radiation chemistry—only the final products could be measured. The rapid advance of radiation chemistry since the advent of pulse radiolysis[50] is well known. Free-radical reactions in the solid state can be followed to some extent by electron spin resonance (ESR). The spin of the unpaired electron can be observed and a reasonable idea of the identities and concentrations of radicals present in a sample obtained.[51]

The main radical species trapped initially in vinyl compounds after irradiation at low temperatures is

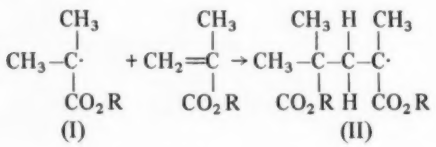
formed by the addition of a hydrogen atom to the double bond.[52] In the methacrylates this is



(I)

Radical I should give an ESR spectrum of seven lines, separated by 23 gauss resulting from equal interaction of the unpaired electron with the six β protons of the methyl groups.[53] A typical spectrum for gamma-irradiated barium methacrylate dihydrate is shown in Fig. 4.

Polymerization of methacrylates by a free-radical mechanism must occur by the reaction



(I) (II)

The propagating radical (II) should give a different ESR spectrum from the initial radical (I). This is found to be nine lines of alternating intensity, separated by 11.5 gauss, as shown in Fig. 5. A major controversy raged for many years over the interpretation of this spectrum, but it has now been fully explained.[53-55]

The change in the ESR spectrum is observed on warming gamma-irradiated methacrylates and is a strong indication that the solid-state polymerization occurs by a free-radical mechanism. However, the ESR spectrum gives no indication of the length of the propagating chain, i.e., it does not distinguish between a dimer and a long chain. This is illustrated by the case of methyl methacrylate, where the change in the ESR spectrum is observed in the solid, but no polymer is produced.

Differential Thermal Analysis

Temperature changes have been used to observe phase transitions in unirradiated solid monomers[56] and the onset of polymerization (strongly exothermic for vinyl monomers) in irradiated monomers.[27] In many monomers polymerization occurs near solid–solid phase changes, indicating the importance of the increased mobility of the monomer. Sometimes the polymerization may be very rapid, e.g., frozen molecu-

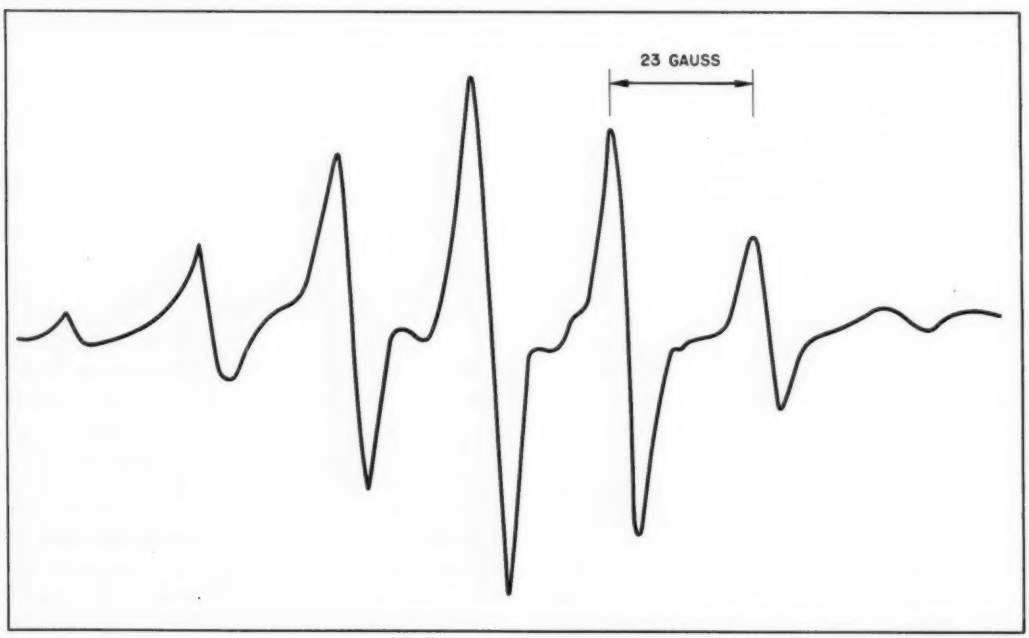


Fig. 4 Electron-spin-resonance spectrum of gamma-irradiated barium methacrylate dihydrate at −196°C.

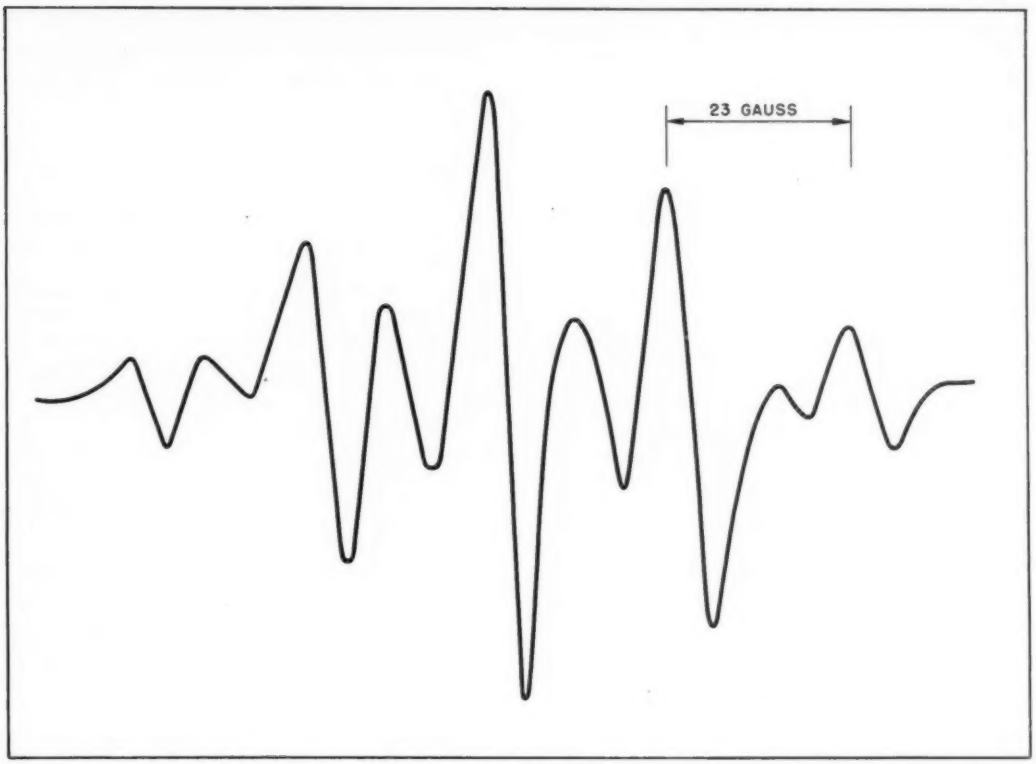


Fig. 5 Electron-spin-resonance spectrum of methacrylate propagating radical in barium methacrylate dihydrate.

lar chains of vinyl monomers and initiators polymerize explosively on warming to about −180°C, where rearrangement of the matrix occurs.[57]

The increased mobility at phase changes can also be observed by ESR. Radicals in gamma-irradiated cyclohexanol disappear near −100°C, where there is a phase change.[58] Methacrylate radicals in gamma-irradiated octadecyl methacrylate are stable up to −35°C, where a phase change occurs and they either polymerize or combine.[59] (MG)

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New Zealand Experience with a ^{60}Co Sterilization Unit

By H. B. Rainey*

Abstract: *The New Zealand ^{60}Co irradiation plant for sterilizing medical supplies is described, together with the bases for its establishment.*

At the time of its commissioning in August 1966, the ^{60}Co irradiation unit in Upper Hutt, New Zealand, was unique in that, although the idea of the plant originated in the organization's needs, it was finally justified and established as a service unit to other companies. Built for the Tasman Vaccine Laboratory by Atomic Energy of Canada Limited (AECL), the principal load for the machine was, and still is, a series of commercially prepared dressing packs for use within the New Zealand hospital system. An increasing range of medical disposables is now being manufactured in this country, and the presence of the unit should ensure that most medical disposables are readily available here; but at present the profitable functioning of the unit is dependent on the dressing packs.

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History

Tasman Vaccine Laboratory was initially concerned solely with the veterinary field, and the need to sterilize plastic syringes for the administration of vaccines was the starting point of the gamma-irradiation project. The acquisition of an ethylene oxide plant was considered, but it soon became obvious that gamma irradiation of properly packed articles was the method of choice.[1] It was equally obvious that no irradiation plant, however small, could profitably be supported by the relatively small volume of packs envisaged.[2] Therefore the possibility of using a unit for medical sterilization purposes was investigated.

The public hospital system in New Zealand is by far the largest user of sterilized disposables in the

country. The hospitals are financed entirely from government funds, and every New Zealander, each of whom pays New Zealand 7.5 cents Social Security tax on every $1 of taxable income, is, together with his family, entitled to medical care of every type without any contributory payment. Although private hospitals exist, and although they receive a 30% subsidy from the state, probably 75 to 80% of all patients hospitalized receive their care in a public hospital.

At the time consideration was being given to the establishment of an irradiation unit, a committee of the Board of Health, the Committee on Sterilisation Procedures, was investigating the standards of hospital sterilization and quite independently stated[3] that a move toward medical disposables and sterilization of bulk items by gamma irradiation would be in the interests of everyone. When it became known that Tasman Vaccine Laboratory was considering such a move, a firm recommendation was made that the hospitals should use such a facility as much as was practicable. Since dressing packs constituted the bulkiest part of the hospital load, an attempt was made to transfer this load to the ^{60}Co unit. Throughout the 37 public hospitals in the country, a total of 247 "standard" packs were used. Often these differed only by a mere half inch in the length or width of the dressing pad; the achievement in reducing these to seven acceptable standard packs (Table 1) was indeed a memorable one. These packs are contracted for annually. All agree that the method of sterilization plus the long-life packs represent a major advance, and realistic costing of conventional hospital sterilization services is gradually convincing any unbelievers.

When a committee of the stature of the Committee on Sterilisation Procedures recommends the introduction of a method of sterilization, it is relatively easy to apply this to an integrated hospital system such as exists in New Zealand. Acceptance is gradually being won, and with it comes a growing tendency to turn to the Tasman Vaccine Laboratory for advice on sterilization. The large storage area available in the plant allows hospitals to use the laboratory's space rather than their own, and one practical result has been the elimination of 10 thousand sq ft of storage space in one new proposed hospital building. With a sound base provided by the dressing packs, Tasman Laboratory is now able to turn its attention to other disposable medical products, many of which are in wide use and some of which are newcomers to the disposable field. With a small country like New Zealand, which has a high standard of medical care available to the whole population, the opportunity exists for the ^{60}Co irradiator to play a significant role in the field of medical disposables and their sterilization.

Table 1 Standard Dressing Packs and Contents as Used in New Zealand Hospitals

1.		Perineal toilet pack
	1	Repellant trolley cover, 20 by 20 in.
	2	Pairs disposable forceps
	10	Cotton wool swabs
	1	Gauze square, 3 by 3 in.
	1	Maternity pad
	1	Disposable diaper, 24 by 16 in.
	1	Plastic tray, 5 by 4½ in., depth ¾ in., divided ⅓ and ⅔
2.		Maternity shower pack
	3	Tissue toilets, 2 ply, 8½ by 5 in., folded once
	3	Paper towels, 16 by 11 in., folded short side to short side
	1	Maternity pad, 9 by 3 in.
3.		Large dressing pack
	1	Plastic trolley cover, 20 by 20 in.
	1	Plastic tray, as for perineal pack
	1	Absorbent dressing guard, 22 by 16 in.
	10	Cotton wool swabs
	3	Gauze squares, 3 by 3 in., 12 ply
	1	Absorbent combine dressing, 9 by 8 in.
	3	Pairs disposable forceps
4,5.		Medium and small dressing packs
		Same as for the large dressing pack but with smaller quantities
6.		Casualty dressing pack
	1	Plastic trolley cover
	1	Plastic tray
	4	Cotton wool swabs
	1	Absorbent combine dressing, 2 by 2 in.
	3	Pairs disposable forceps
7.		District nurses pack
	1	Plastic trolley cover
	1	Plastic tray
	1	Absorbent dressing guard, 22 by 16 in.
	4	Cotton wool swabs
	4	Gauze squares, 2 by 2 in., 12 ply
	1	Absorbent combine dressing, 9 by 4½ in.
	1	Paper towel
	3	Pairs disposable forceps

Facility Description

The facility is designed to irradiate boxes of medical products to a minimum dose level of 2.5 Mrads of ionizing radiation from a ^{60}Co source. The capacity

of the plant is 750 thousand cu ft/year with a 1-Mc source. The cobalt strength at commissioning, in August 1966, was 154 thousand curies and was supplemented a year later by 50 thousand curies, giving a present strength of 184 thousand curies. The plant is designed to operate at 95% efficiency, assuming that the prescribed program of maintenance is followed assiduously.

The irradiator (Fig. 1) is designed so that the product is taken to the irradiation cell through a maze, the source being surrounded by a concrete biological shield. The product, packed in a standard cardboard box 11 in. wide by 15.25 in. long by 16.9 in. high overall, is carried in an aluminum carrier on a ceiling rail. The boxes of these dimensions are normally handled in pairs, two pairs to a carrier, but a single box 33.8 in. high could quite easily be carried. Each small box has a volume of 1.64 cu ft, weighs 20.5 lb when loaded, and has a product density of 0.2 g/cm^3. There are no problems, however, with densities up to 0.35 g/cm^3. The maximum/minimum dose ratio at a density of 0.2 g/cm^3 is 1.2:1.0.

Building

The building housing the irradiator consists basically of a concrete biological shield surrounding the source. The shield, which is above ground, is constructed of standard-density concrete. The walls are 5 ft thick and are designed to reduce levels of radiation outside to not more than 0.25 mr/hr at the surface with a full source load of 1 million curies when the source is in the operating position. The roof, which is 4 ft thick, reduces the level of radiation to not more than 100 mr/hr. A hatch directly over the pool allows replenishment of the source. The construction is such that personnel will not receive doses greater than 3 mr/week in external areas and 10 mr/week in internal areas.

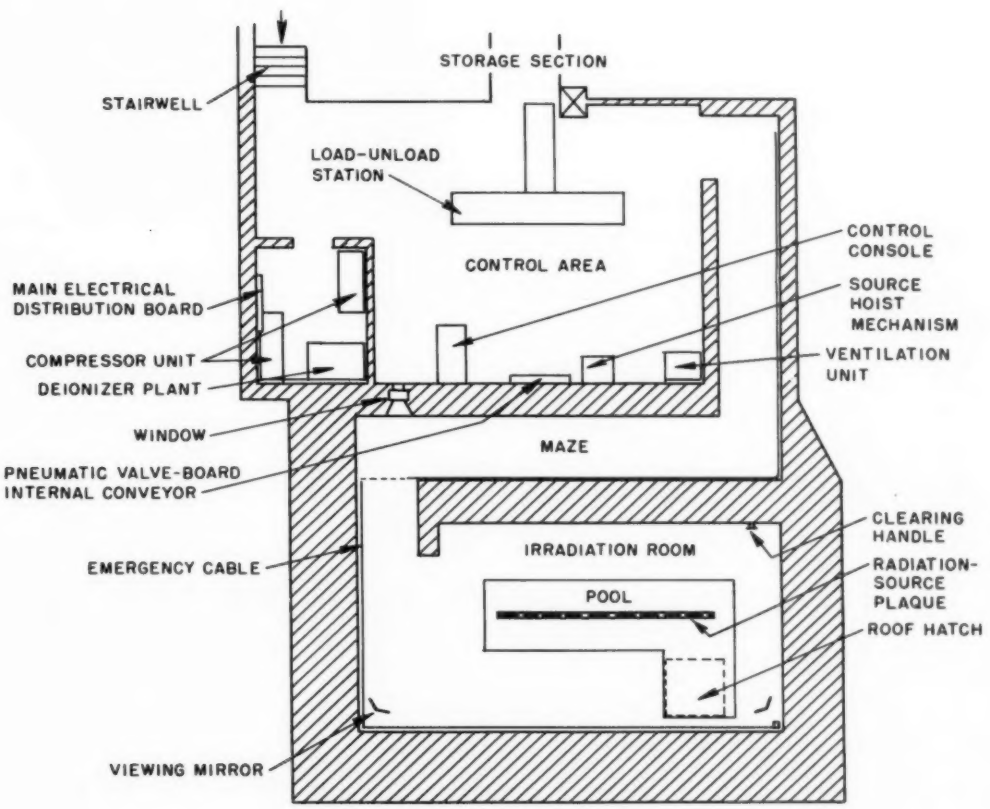

Fig. 1 Layout of New Zealand ^{60}Co irradiator.

The biological shield is constructed over a 26-ft-deep well into which the source descends in emergency or when not functioning. The pool walls are of 2-ft-thick concrete with an outer proof lining and an inner industrial tile lining.

Conveyor System

The conveyor system leads into, and away from, the load–unload point. The loading station at the head of the "in" conveyor consists of a table with a carton gage that will not accept any offsize or irregular cartons. Together with the unloading of the discharge conveyor, this is the only manual operation in the process. The storage or "in" conveyor is designed to allow 24-hr operation at the present source strength but can be increased, as can the discharge or "out" conveyor, to accommodate any number of cartons up to the maximum source capacity.

The load–unload station transfers an unirradiated pair of cartons from the "in" conveyor to the top half of the aluminum carrier, at the same time discharging a sterile pair of cartons from the bottom of the carrier to the "out" conveyor and transferring the original top pair to the bottom position. This maneuver is performed by a series of vertical and horizontal transfer devices motivated by pneumatic cylinders. The net result is that each pair of cartons travels twice around the source, ensuring an even distribution of radiation in each carton. The conveyor for the carriers, a power-driven overhead monorail type, takes the carriers through the maze, around the source, and back to the load–unload station via the maze.

The carriers are arranged in the irradiation cell in 6 rows, 14 carriers to each row (Figs. 2 and 3). With 4 cartons, i.e., 2 pairs, to each carrier, this makes a total of 336 cartons in the cell at any one time.

Fig. 2 Carriers with cartons inside radiation cell. Rows 5 and 6 contain only eight carriers in this picture, which was taken during construction.

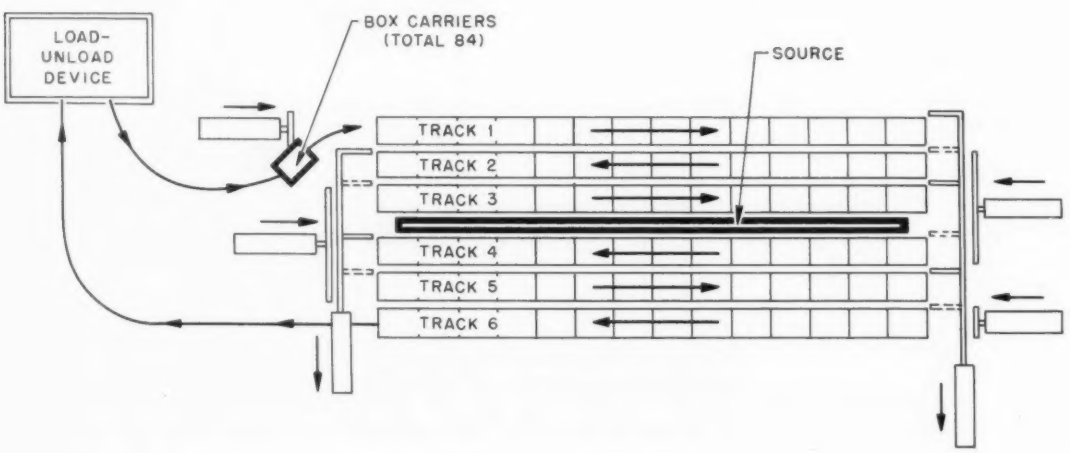

Fig. 3 Operating sequence of internal conveyor.

The carriers are moved around the source by regular intermittent movements and, in acquiring the calculated radiation dose, occupy 168 different positions in relation to the source. The movements are controlled by a master timer, the time setting being determined by the source strength and the dose required.

Cobalt Source

The ^{60}Co is in the form of small pellets, 1.0 by 1.0 mm, which are doubly encapsulated in stainless-steel tubing whose ends are welded. These pencils are loaded into the source frame, which is 4 ft 8 in. high by 17 ft 0 in. long. The rack consists of 20 modules in two rows of 10, each module containing 42 pencils. The pencils are numbered serially. The source movement is guided by two stainless-steel cables, one at each end of the frame. The frame itself is raised and lowered by another set of stainless-steel cables leading to a hoist mechanism in the load–unload area.

Control Console

The control console is located in the control area. It contains a radiation monitor section and a section for the electrical apparatus and relays that regulate the automatic operation of the machine (Fig. 4).

Safety Mechanisms

The movements of carriers and cartons at the load–unload area and within the radiation cell are carried out by pneumatic cylinders. In the event of a power failure, the source rack will be lowered automatically. The unit is constructed less than half a mile from a major earthquake fault, and an earthquake greater than force 4 on the Richter scale will activate a monitor that cuts off all power to the irradiator. The rack will then automatically drop into the pool. If the hoist cables slacken owing to an obstruction to the rack in its descent, a sensing switch will lock the hoist until the cable is tightened. If the obstruction persists, the source will move down intermittently. The machine will shut down automatically if any part of the conveying system does not complete a cycle within a prescribed time.

A sensing device in the maze will detect distorted cartons but will allow a normal carton to pass through without actuating it.

There is also a device in the cell for detecting a temperature greater than 100°F, in which case the

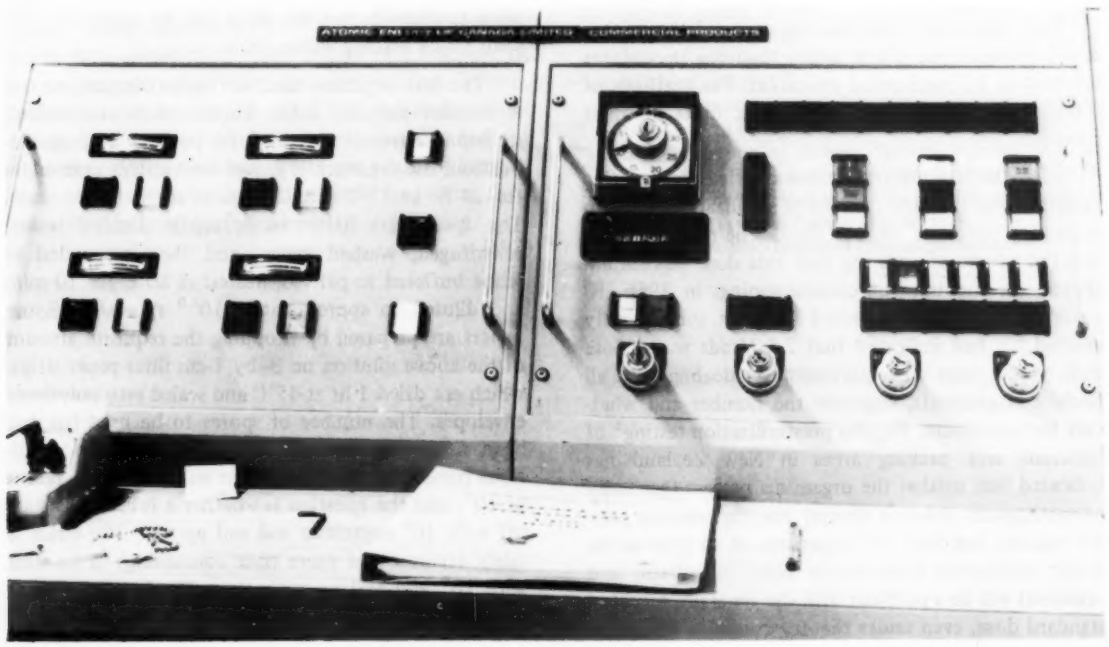

Fig. 4 Control console of New Zealand ^{60}Co irradiator.

source is lowered and the machine stopped. If the level of the water in the pool drops below a predetermined level, a light on the control console is activated, warning the operator. When the air pressure drops below a certain level, the conveyor system is shut down and the source drops.

The start handle is in the cell and must be pulled by the operator to start the machine. This means, of course, that he must walk into the cell before starting the machine and would see any person who might be there. The handle has a 60-sec delay after being actuated, but the source rack will still not rise until the door dividing the maze from the control area is shut and locked. The source can be lowered by pulling an emergency cable and by emergency stop buttons on the control console and load–unload subpanel. A remote signal can be sent to the local Fire Brigade Station, which is manned 24 hr/day, to warn of an unintended shutdown.

The irradiation area is provided with ventilation apparatus that changes the air in the cell every 4 min, thus keeping ozone below the accepted maximum level of 0.1 ppm.

Plant Operation

This subsection describes the uses and the day-to-day control of the plant rather than the procedures involved in its mechanical operation. The methods of control are based on those used in units in other countries.

After the hospital system was satisfied that ionizing radiation was the ideal method of sterilization, the two remaining problems were to decide the radiation dose and the means of ensuring that this dose was administered. At the time of commissioning, in 1966, 10 years' experience in the United Kingdom, subsequently verified,[4-7] had indicated that 2.5 Mrads was a dose that would meet all requirements in dealing with all likely contaminants, whatever the number and whatever the conditions. Regular presterilization testing[8] of materials and packing areas in New Zealand has indicated that neither the organisms present (generally nonpathogenic airborne spores) nor the number present (usually less than 10^3 organisms of all types on an article containing components made of plastic and cellulose) will be a problem with the generally accepted standard dose, even under the dry conditions existing. Checks on the type of organisms present in the packing areas and on the personnel who do the packing have never indicated organisms that were not more than adequately destroyed by 2.5 Mrads.

It can be argued that, with a reliable clock and a source of known strength, no check on the dose rate is needed once initial work has been done to determine the dose for any given length of time in the cell. This would be feasible if product of constant density was continuously handled. Although increasing density does not necessitate a linear increase in radiation dose,[9] there is often sufficient shielding by a carton with a density >0.2 g/cm^3 (i.e., 20.50 lb per standard carton) to reduce the dose level absorbed below 2.5 Mrads. Experience helps in positioning cartons of higher than average density within the cell, but even so it is important to know the administered dose. The two methods of control used are physical, with a red acrylic (Perspex) disk, and bacteriological, with a relatively radiation-resistant organism.

The acrylic-disk system measures the accumulated dose to ±5% in a dose range of 0.5 to 5.0 Mrads. Radiation increases the optical density of the red acrylic pellet, and the dose is read off on a dosimeter manufactured by AECL, which has been set to a zero reading with an unirradiated disk. The disks are 0.375 in. in diameter and 0.373 in. thick. The acrylic is stained with a red dye whose color fades on storing after irradiation, but the error can be limited to less than 1% by reading within 48 hr.

The test organism used for bacteriological control is *Bacillus pumilus* E601, known to be a relatively radiation-resistant organism. To prepare a spore suspension, the organism is grown on nutrient agar slopes for 24 hr at 37°C plus 2 days at room temperature. The spores are harvested in sterile distilled water, centrifuged, washed twice, and then suspended in saline buffered to *p*H 7.0, heated at 80°C for 10 min, and diluted to approximately 10^{10} spores/ml. Spore papers are prepared by dropping the requisite amount of the above dilution on 3- by 1-cm filter paper strips, which are dried 1 hr at 45°C and sealed into polythene envelopes. The number of spores to be used has not been finally decided. At 2.5 Mrads under the conditions prevailing, *B. pumilus* has an inactivation factor of 10^7, and the question is whether it is better to start off with 10^8 organisms and end up with 10 (which is fairly accurate but more time consuming) or to start with 10^5 and end up with 10^{-2}. The latter system is used at present.

Each spore paper is inserted in an envelope with a red acrylic disk, and one envelope is attached to the

top center of both the upper and lower cartons of one test pair for each 50 pairs of cartons, meaning that at any one time there are three control pairs of cartons in the cell. When the test cartons emerge from the cell, the disks are read in the dosimeter and the spore papers are incubated, with nonirradiated spore papers as controls, 5 days at 37°C. If both the physical and bacteriological test results are satisfactory, the cartons are released for distribution. A satisfactory dosimeter test is defined as one in which the readings of 48 consecutive dosimetry disks are not less than 2.5 Mrads, not more than 5 disks read below 2.35 Mrads, and not more than 1 disk reads below 2.25 Mrads. The dosimeter was calibrated at commissioning by AECL, and dosimeter and disks are checked quarterly by the Institute of Nuclear Sciences of the Department of Scientific and Industrial Research.

If product of uniform density is being processed, one quality-control envelope in the cell at any one time would be sufficient. However, with a unit such as the one at the Tasman Vaccine Laboratory, which irradiates cartons from many sources, differences in density with consequent shielding by heavier cartons necessitate a greater number of quality-controlled cartons, and three is the number chosen. Cartons are frequently irradiated for firms that are competitive in the veterinary field and may not wish to divulge the contents. Under these circumstances the only responsibility taken is to ensure the administration of 2.5 Mrads. If, as is generally the case, the contents are known, additional assurance is given that the irradiated product is suitable for the intended purpose. To this end our chemistry department has carried out a large volume of trial work on materials with a view to testing their suitability for, and performance after, irradiation.

(MG)

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Section V

Isotopes and Radiation Technology

Isotopic Heat and Power Development

Isotopic Power in Europe–II*

By Jerome G. Morse†

Abstract: *An increase in active isotopic power source programs in Europe from 17 to 27 in one year is indicative of growing interest. This paper includes comments on specific national and international programs, on future growth, and on terrestrial and space safety.*

In Europe, isotopic power source technology is a progressive and continually expanding process. Twenty-seven active programs exist in Europe today, as opposed to the 17 reported 1 year ago.[1] All programs identified in this paper‡ are aimed at the development of devices that produce electricity—indirectly through a preliminary conversion to heat—from the kinetic energy of the products of radioactive decay. Most of the devices are prototypes for test and evaluation, with mission identity to be established in the future. The hardware effort is concurrent with the research and development in the areas of fuel and energy conversion.

Expansion in the European isotopic power source program may be attributed to:

1. Increased investment in research and development. This is noticeable on a national scale, but more importantly, a spirit of intergovernment cooperation in technology exchange has been stimulated by many organizations, particularly ENEA, thus reducing considerably the developmental overlap. Increased cooperation with the United States would be welcomed and, in the writer's opinion, would offer noticeable advantages to all parties concerned.

2. The fact that isotopic power represents a facet of the nuclear field wherein technical growth and operational hardware may be realized at relatively modest investment.

Generators for space missions continue to be funded, but Europe's acceptance of a rather limited role in space exploration during the next decade precludes a large expansion and investment in generator technology. Applications are seen to be restricted to earth orbit (communications and meteorology) and scientific probes with a limited number of units to be launched. Expansion has occurred, however, in terrestrial devices, especially for those to be used in undersea roles, for it is in this environment that isotopic power can be clearly cost competitive with conventional energy sources.

Britain continues as the hardware leader, whereas Germany has emerged as the strongest in research and development. Fuel processing and production are still within the province of the government laboratories, but specific companies can now be identified as suppliers of energy-conversion components; for ex-

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‡The international organizations, most of the national organizations, and some of the companies are abbreviated in text. The abbreviations are explained in Table 1, where they are arranged in alphabetical order. Nonabbreviated organizations and companies are also included in Table 1.

Table 1 Abbreviations, Acronyms, and Names of Organizations and Companies Active in Isotopic Power Technology

Abbreviation	Organization or company	Country	Location
AEG	Allgemeine Elektricitats-Gesellschaft	Germany	Hamburg, Germany
AERE	Atomic Energy Research Establishment, UKAEA	England	Harwell, Berks, England
ALCATEL	Société Alsacienne de Constructions Atomiques de Télécommunications et d'Electronique	France	Paris, France
ARPA	Advanced Research Projects Agency	United States	Washington, D. C.
	Bolkow (see Ludwig Bolkow Entwicklungen KG)		
BBC	Brown, Boveri & Cie.	Germany	Heidelberg, Germany
CEA	Commissariat à l'Energie Atomique	France	Paris, France
CEN	Centre d'Etude de l'Energie Nucléaire	Belgium	Brussels, Belgium
CNES	Centre Nationale d'Etudes Spatiales	France	Paris, France
CSF	Cie. Générale de Télégraphie sans Fil	France	Paris, France
DRME	Directions des Recherches et Moyens d'Essais-Ministére des Armes	France	Paris, France
ELDO	European Launching Device Organization or European Launcher Development Organization	Belgium, France, Germany (Federal Republic), Italy, Netherlands, United Kingdom, Australia	Delft, Netherlands*
ENEA	European Nuclear Energy Agency, OECD	Austria, Belgium, Denmark, France, Germany (Federal Republic), Greece, Iceland, Ireland, Italy, Luxembourg, Netherlands, Norway, Portugal, Spain, Sweden, Switzerland, Turkey, and United Kingdom with Canada, Japan, and the United States of America as associate members	Paris, France*
EURATOM	European Atomic Energy Community	Belgium, France, Germany (Federal Republic), Italy, Luxembourg, and Netherlands	Brussels, Belgium* Joint Nuclear Research Center, Ispra, Italy†
EUROSPACE	Groupement Industriel Europeen d'Etudes Spatiales	Belgium, France, Israel, Denmark, Germany (Federal Republic), Italy, Netherlands, Norway, Spain, Sweden, Switzerland, United Kingdom, and important U. S. companies corresponding members	Paris, France*
	Hispano-Martin	France	Bois-Colombes, France
	Hispano-Suiza	France	Bois-Colombes, France
IAEA	International Atomic Energy Agency	93 member countries	Vienna, Austria*
	Junkers & Co. GMBH	Germany	Munich, Germany
	Ludwig Bolkow Entwicklungen KG	Germany	Bavaria, Germany
	Metallurgie Hoboken	Belgium	Antwerp, Belgium
	Mining and Chemical Products Ltd.	England	London, England
OECD	Organization for Economic Cooperation and Development	Governments of the 21 member countries of ENEA (Canada, Japan, and the United States of America full members of OECD) with Finland and Yugoslavia with special status but not full membership	Paris, France*

*Headquarters.
†Manufacturer.

(Table continues on the next page.)

Table 1 (Continued)

Abbreviation	Organization or company	Country	Location
OECD (continued)	Philips N. V.	Netherlands	Eindhoven, Netherlands
	G. V. Planer Ltd.	England	Sunbury-on-Thames, Middlesex, England
	Plessey Co. Ltd.	England	Caswell, Towcester, Northamptonshire, England
RCN	Reactor Centrum Nederland	Netherlands	The Hague, Netherlands
	Siemens	Germany	Munich, Germany
SNECMA	Société Nationale d'Etude et de Construction de Moteurs d'Aviation	France	Suresnes, (Seine), France
UKAEA	United Kingdom Atomic Energy Authority	United Kingdom [Great Britain (England, Scotland, and Wales) and Northern Ireland]	London, England*
USAEC	United States Atomic Energy Commission	United States of America	Washington, D. C.*

*Headquarters.

ample, sources of thermoelectric materials include the following:

Country	Company	Thermoelectric material
Belgium	Metallurgie Hoboken	Bi_2Te_3, PbTe, and GeSi
France	CSF	Bi_2Te_3
	ALCATEL	PbTe
Germany	AEG	GeSi
	Siemens	GeSi
United Kingdom	Mining and Chemical Products Ltd.	Bi_2Te_3 and PbTe
	G. V. Planer Ltd.	PbTe
	Plessey Co. Ltd.	GeSi

Generators are not yet commercially available, as they now are in the United States.*

The purpose of this paper is to note program status where data are available, to generalize on future growth, and to discuss terrestrial and space safety programs.

*On Aug. 31, 1967, *Nucleonics Week* announced that Submarine Cables Ltd., London, U. K., is manufacturing 4-watt(e) isotopic power sources for commercial sale. The Isotopic Thermoelectric Generators are licensed to Submarine Cables by the UKAEA, which calls its device RIPPLE (*R*adio *i*sotope-*p*owered *p*rolonged-*l*ife *e*quipment).

Commentaries on Programs

EURATOM

The EURATOM in-house efforts are listed in Table 2, and the single contractual program, GETER-1, is noted in Table 3.

Although the fueling date has not yet been announced, EURIG-2 may be the world's first thermoelectric generator powered by ^{170}Tm. The fuel may be obtained from the CEN Mol reactor, Mol, Belgium. A decrease in power output with time can be anticipated with the furnace-brazed PbTe thermoelements or couples operating at the design hot-side temperature. The work appears to be well under way after a slow start and lengthy gestation period.

In addition to proposed application in the EURO-SPACE Program, EURIG-3 will probably be developed to higher power levels [>50 watts(e)]. Further, the planned use of ^{227}Ac is predicated upon the availability of large radium inventories, which have been obtained from former African colonies. The conversion of radium to actinium is in active laboratory development.

Impressive results have been obtained in thermionic research and development (HPTC-1) for both isotope- and reactor-powered systems. Heat-pipe research is another active program.

France

A broad application spectrum of thermoelectric devices is seen in Table 3. There appears to be no

Table 2 EURATOM In-House Efforts

Designation	EURIG-2	EURIG-3	HPTC-1
Use	Demonstration (terrestrial)	Design study (space)	Laboratory prototype (space)
Fuel form	$^{170}Tm_2O_3$	$^{227}Ac_2O_3$	$^{227}Ac_2O_3$
Inventory	40 kilocuries 200 g	57 g	22.5 g
Generator electric power			
BOL,* watts(e)	5	50	50
Open-circuit voltage, volts	5	5	1.0
Size, cm			
Diameter	19	25	5
Height	13	25	25
Design life, years	-	1	1
Weight, kg	90	2	0.4
Status	Test with electric heater	Design	Test
Manufacturer	EURATOM	EURATOM	EURATOM
Type of conversion device	Thermoelectric, PbTe, 30 thermoelements	Thermoelectric, GeSi, 20 thermoelements	Thermionic, Rh emitter, Ni collector
Source of conversion material	-	Metallurgie Hoboken	
Overall generator efficiency, %	6.2	5.9	16
Temperature, °C			
Hot	550	1000†	1777
Cold	100	500†	700
Shield material	Pb	None	None

*Beginning of life. †Calculated.

Table 3 French Thermoelectric Devices

Designation	None	SPIREE	None	MARGUERITE	GETER-1
Use	Demonstration (space)	Prototype (space)	Demonstration (terrestrial)	Beacon (undersea)	Demonstration (terrestrial)
Fuel form	^{210}Po (metal deposited on Pt)	$^{90}SrTiO_3$	$^{90}SrTiO_3$	$^{90}SrTiO_3$	$^{90}SrTiO_3$
Inventory	0.2 kilocurie	230 watts(e) EOL*	15 kilocuries	0.8 kilocurie	0.9 kilocurie
Generator electric power, watts(e)	0.3 BOL†	10 EOL*	~3 BOL†	0.090 BOL†	0.25 BOL†
Open-circuit voltage, volts	1.0	2.1	-	0.235	0.96
Size, cm					
Diameter	6.6	-	-	35	-
Height	8.5	-	-	65	-
Design life, years	-	5	-	-	5
Weight, kg	0.5	8 (without shield)	-	700	
Status	Fueled 6/66	Generator test 1968	-	Test with electric heat	Design
Manufacturer	CSF, CEA	CSF, SNECMA, CNES	ALCATEL	SNECMA, SCF	Hispano-Suiza, EURATOM
Type of conversion device	Thermoelectric, Bi_2Te_3	Thermoelectric, GeSi	Thermoelectric -	Thermoelectric, Bi_2Te_3	Thermoelectric, Bi_2Te_3, 16 thermo-elements
Source of conversion material	CSF	CSF	-	CSF	Metallurgie Hoboken
Overall generator efficiency, %	4.7	4.5	-	1.8	2.46
Temperature, °C					
Hot	200	900	-	200	220
Cold	25	250	-	35	70

*End of life. †Beginning of life.

central sponsoring agency funding developments. The CEA has complete responsibility for fuel work, but the CNES, Navy, etc., award the contracts for generator development, coordinating with the CEA for fuel and fueling. In recent months, DRME—equivalent to ARPA—has attempted to coordinate and plan future developments.

The unnamed ^{210}Po device was intended for laboratory demonstration and, as such, has fully met its objective. To achieve the relatively high voltage (1 volt) at 0.3 watt, it was necessary to use long slender thermoelements with questionable ability to meet the rigors of a launch environment. This "application –extrapolation" is only that of the writer.

SPIREE was redirected in past months and is now primarily a test bed for GeSi thermoelement technology. The decision was predicated on the CNES' low-priority need for long-lived nuclear power supplies before the early or mid-1970's.

The relatively low efficiency of MARGUERITE is attributable to the conductive copper leaf spring in thermal contact between (1) the heat source and the thermoelements and (2) the thermoelements and the outer surface. This appears as excessive conservatism in design in view of the innate structural stability of Bi_2Te_3.

Although early design data on GETER-1 indicate hot-side temperatures of 220°C, test data on Bi_2Te_3 elements have been reported at 300°C for more than 10,000 hr. Under the test-data conditions, bonded couples have demonstrated power outputs stable to ~1% with an increase in power performance of 50% over the early-design-data lower values.

In addition to the above programs, funded programs may soon be announced covering:

1. A mixed fission-product (or ^{90}Sr-)-fueled dynamic system, probably a Stirling-cycle engine, in the range of 1 to 10 kw for a small manned submersible. This program is being stimulated by the Navy and DRME.

2. A 5-watt ^{90}Sr-fueled thermoelectric device (PbTe thermoelements) for undersea use. This program is also being stimulated by the Navy and DRME.

3. A Poodle-type thruster using ^{144}Ce. Initiated by the CNES, the program is now in parametric evaluation with alternative fuels, such as ^{210}Po, being considered.

Germany

The programs identified in Table 4 reflect a decided space orientation. Impressive data have been reported on testing of GeSi bonded thermoelements at about 1000°C in air and in vacuum.[2] Unfortunately, development of fuel-containment materials capable of operating at the design temperatures has slowed advancement to the operational hardware stage. The programs appear to uniformly favor $SrTiO_3$ as the space fuel, but there is little capability in Germany to supply fuel in the near future. Considerable sophistication in generator design is in evidence.

Netherlands and ENEA

Both programs (Table 5) are in a study phase, and both anticipate using the Stirling cycle for energy conversion. With respect to the latter, the Philips work continues to be impressive and engines ranging from a few to 100 kw are under life test, with a design goal of 10,000 hr. Philips and RCN are now teamed in a combined preliminary systems and market-potential study.

Apparently either the Netherlands or ENEA program or the French counterpart, if given a firm funding base, can well leapfrog the United States in this type of power supply.

United Kingdom

As Table 6 shows, the UKAEA has clearly embarked on a national program to demonstrate the capabilities of isotopic power. Concentrating on terrestrial applications, the hardware effort has been quite successful. Designs have been practical and conservative, and the intent is to create a market for the hardware in Europe and elsewhere. To date, four units have been, or are being, loaned for field test to familiarize the users with the device, and it is anticipated that program growth (and U. K. industrial involvement) will follow the test phase.

Union of Soviet Socialist Republics

The national programs are probably more extensive than indicated in Table 7. Further, it has been noted that BETA II-type hardware in limited amounts is being offered for sale at international conferences.

Fuel

For the sake of completeness, Table 8 is included, but it represents no new data over those previously reported.[1]

Safety

All terrestrial generators are being designed in accordance with the IAEA regulations concerning the

Table 4 German Program

Designation	None	None	None*	None
Use	Satellite	Satellite	Satellite	Development (space)
Fuel form	Undefined	$^{90}SrTiO_3$	$^{90}SrTiO_3$	Undefined (50 to 100 watts/cm^2 surface thermal flux using heat pipes)
Inventory, kilocuries	-	-	-	-
Generator electric power				
BOL,† watts(e)	50	125	500 to 3000	200 to 250
Open-circuit voltage, volts	-			
Size, cm				
Diameter	-	-	-	-
Height	-	-	-	-
Design life, years	5	-	1	-
Weight,‡ kg/watt	0.4	0.5	100	
Status	Generator in performance test	Generator fabricated 7/66	-	
Manufacturer	AEG	Bolkow	Junkers (Space Ministry)	BBC, EURATOM
Type of conversion device	Thermoelectric, GeSi, 72 thermo-elements	Thermoelectric, GeSi	Turboelectric, Rankine cycle, Hg working fluid	Thermionic, Cs vapor, Mo emitter and collector
Source of conversion material	AEG	Siemens	Junkers	BBC
Overall generator efficiency, %	-	~6	~12	17.4§
Temperature, °C				
Hot	1000	827	-	1700
Cold	-	327	-	-

*Terminated fall 1966—funding problems. †Beginning of life. ‡Without shield. §Measured.

Table 5 Netherlands and ENEA Programs

	Netherlands	ENEA	
Designation	None	MASTODON-1	MASTODON-2
Use	Prototype (terrestrial)	Prototype (undersea stationary)	Prototype (undersea submersible propulsion)
Fuel form	^{90}Sr or ^{144}Ce	$^{90}SrTiO_3$	$^{144}CeO_2$
Inventory, kilocuries	-	-	-
Generator electric power			
BOL,* kw(e)	1 to 10	1 to 10	10 to 100
Size, cm			
Diameter	-	-	-
Height	-	-	-
Design life, years	1	1	1
Weight, kg	-	-	-
Status	Power-system study under way; engine under life test	Engineering study	Engineering study
Manufacturer	Philips N. V. and RCN	†	†
Type of conversion device	Stirling cycle	Stirling cycle	Stirling cycle
Source of conversion material	Philips N. V.	Philips N. V.	Philips N. V.
Overall generator efficiency, %	~40	~40	~40
Temperature, °C			
Hot	700		
Cold	30		

*Beginning of life.

†Possible task breakdown:

Fuel development and heat exchangers—France and United Kingdom. Safety—Spain.

Conversion—Netherlands. System integration—Netherlands.

Table 6 United Kingdom Program

Designation	RIPPLE I and II	RIPPLE III and IV	RIPPLE V to VII	RIPPLE VIII to XII
Use	Demonstration (terrestrial)	Cable repeater (submarine)	Navigation light (marine)	Variable (terrestrial)
Fuel form	$^{90}SrTiO_3$	$^{90}SrTiO_3$	$^{90}SrTiO_3$	$^{90}SrTiO_3$
Inventory, kilocuries	0.7	4.6	-	-
Generator electric power				
BOL,* watts(e)	0.078	0.75	1.0	5 to 100
Open-circuit voltage, volts	0.4	-	-	-
Size, cm				
Diameter	22	-	-	-
Height	23	-	-	-
Design life, years	10	5	5	-
Weight, kg	230	275	-	-
Status	Continuous operation since 3/65	Generators fueled and awaiting field test	Generators fueled and awaiting field test†	Stages of design or fabrication
Manufacturer	UKAEA, AERE	UKAEA, AERE	UKAEA, AERE	UKAEA, AERE
Type of conversion device	Thermoelectric, Bi_2Te_3, 18 thermoelements	Thermoelectric, Bi_2Te_3	Thermoelectric, Bi_2Te_3	Thermoelectric, PbTe
Source of conversion material	Mining and Chemical Products Ltd.	Mining and Chemical Products Ltd.	-	G. V. Planer Ltd.
Overall generator efficiency, %	1.8	2.4	-	-
Temperature, °C				
Hot	180	-	-	-
Cold	40	-	-	-

*Beginning of life.

†Testing program using RIPPLE hardware planned by: Trinity House (V), Coast Guards of Sweden (VI) and Denmark (VII), and U. S. Navy Facilities Command.

Table 7 Union of Soviet Socialist Republics Program

Designation	BETA I	BETA II	PROTO V*	PROTO VI*	PROTO VII*
Use	Automatic weather station	Automatic weather station	Demonstration (space)	Demonstration (space)	Demonstration (space)
Fuel form	^{144}Ce molybdate	$^{90}SrTiO_3$	^{210}Po metal	^{210}Po metal	^{210}Po metal
Inventory, kilocuries	17.5	30	10	10	7.7
Generator electric power					
BOL,† watts(e)	5.6	5 to 7	10.2	10.9	5.8
Open-circuit voltage, volts	3.5	3.5 to 4.5	3.9	3.6	1.4
Size, cm					
Diameter	-	35	19.5	19.6	19.0
Height	-	40	12.8	12.8	21.0
Design life, years	-	10	-	-	-
Weight, kg	400	500	3.1 (without isotope capsule)	2.7 (without isotope capsule)	2.8 (without isotope capsule)
Status	In operation since 1964	Experimental operation	In operation since 1965	In operation since 1965	In operation since 1965
Manufacturer	-	-	-	-	-
Type of conversion device	Thermoelectric, Bi_2Te_3	Thermoelectric -	Thermoelectric, GeSi, 8 thermo-elements	Thermoelectric, GeSi, 8 thermo-elements	Thermoelectric, GeSi, 8 thermo-elements
Source of conversion material	-	-	-	-	-
Overall generator efficiency, %	4	3.5	3.2	3.4	2.4
Temperature, °C					
Hot	300	-	860	840	760
Cold	20	-	240	250	230
Shield material	Pb	-	-	-	-

*Cosmos series of satellites. †Beginning of life.

Table 8 Radioisotope Production Capability in Europe[1]

Radioisotope	Potential annual production kw (thermal)	Year beginning*	Radioisotope preparation	Compound
^{90}Sr	120	1970	Fission product	$SrTiO_3$
^{137}Cs	80	1970	Fission product	Polyglass
^{144}Ce	1000	1970	Fission product	CeO_2
^{147}Pm	15	1970	Fission product	Pm_2O_3
^{210}Po	100	1970	Reactor activation	Metal, rare-earth polonides
^{227}Ac	2	1968	Reactor activation	Ac_2O_3
^{238}Pu	1	1968	Reactor activation†	PuO_2

*If needs justify startup.
†By 1970, 20 kg of ^{237}Np and, after 1970, 10 kg of ^{237}Np per year will be available from the United Kingdom.[4]

transport of radioactive material. Further, the specific application will define the adequacy of design safety constraints, which will be evaluated by the atomic energy organization within whose jurisdiction the application lies. The IAEA exercised leadership in this area in July 1966 by convening a meeting to standardize terrestrial safety criteria—realistically and to the maximum extent possible.

For reasons not identified, the IAEA elected not to extend the above activities to space. Safety considerations in space generator design and development have been noticeably absent. The majority of systems have anticipated the use of $SrTiO_3$, allowing sufficient volume in the generator for the necessary thermal flux to be provided by the encapsulated fuel. Consideration of the ultimate fate of the fuel and its influence on generator design has been wanting. This may be attributed in large part to both lack of experience in Europe and lack of design information from those with the experience using other fuels, namely, the United States and Union of Soviet Socialist Republics. To fill this void, ELDO has taken the lead recently in space safety. A group of scientists representing the nations of Western Europe and the technical disciplines involved has been meeting to define the boundary conditions of the design problem. Studies have been initiated to define the constraints imposed by typical missions and to identify the analytical and testing programs that must follow. This is a most significant start.

Information Exchange

With the awareness of an expanding body of technology engendered in a number of small but growing programs, steps have been taken to exchange information to a maximum extent. Two events are noteworthy:

1. In September 1966, UKAEA and ENEA cooperated in a conference to bring the generator designers together with the potential users. The meeting proved to be very useful for both groups of participants. All papers have been published in a single volume[3] by OECD. The success of the first conference will undoubtedly lead to others.

2. ENEA, with CEA, established an experimental "Isotopic Generator Information Centre," which issues a quarterly newsletter that contains technical papers, news items related to the field, and a bibliography of recent articles published in Europe and elsewhere. This is a highly commendable and useful effort that will further information exchange.

Conclusions

Isotopic power source technology in Europe is definitely expanding. Although the actual funding is not known to the writer, it is likely that the present rate is much less than that of the U. S. program. The effort is considerable, however, and the impact will be greater by virtue of the leadership of international organizations to exchange data. The latter is not a *fait accompli* since programs are duplicated on a national basis.

Progress has been made in the safety area as it has been recognized in Europe that accidents in the terrestrial or space environments become everyone's problem. Safeguards, analogous to those established for reactors, are being identified.

United States technology continues to lead, particularly in static energy conversion, but the progress made in Europe with modules of Bi_2Te_3 and GeSi alloys is worthy of watching closely. The one area in which there is an opportunity to lead the United States

is in the coupling of isotopic energy sources with Stirling-cycle engines for undersea use. (JEC)

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Section
VI
Isotopes and Radiation Technology

Applications in Life Sciences

Sixty-Five Years of Medical Radioisotope Scanning* (Which Is Clinically Really Only About 5 Years Old)

By Marshall Brucer, M.D.

{Editor's Note: Dr. Brucer decided against updating this paper to 1968 since the situation with regard to radioisotope scanning has changed very little. "The Anger Camera" by Joan E. Carden [*Isotopes and Radiation Technology*, 5(1): 45-53 (Fall 1967)] describes the recent status of one important scanning device.}

The routine clinical use of medical radioisotope scanning actually goes back only to the early 1960's. The first 10 years of scanning was spent in a very necessary, frustrating, and sometimes unproductive instrumentation development. A primitive kind of scanning was done with natural radioactive materials long before the development of artificial radioactivity; thus artificial radioisotopes are not the historic beginning of scanning. However, scanning is unthinkable without a highly sensitive method of detection. Nor could radioisotope localization be even considered without a prior development of the concept of the field-of-vision of the detector.

It is still too early to write a definitive history of scanning, because everything we consider a technical triumph today appears to be an oversimplified version of a still greater technical triumph tomorrow. Naturally, just as interesting a story is connected with the development of the scanning radiopharmaceuticals. As in the fields of diagnostic radiology or anatomical pathology, the really informative history will concern the physician's ability to interpret his observations.

Today, most of us are still primarily concerned with machinery. The piles of iron that sit in the corners of so many diagnostic laboratories, and either do or do not move back and forth across the patient to produce a picture of a localization, usually contain a small (3-, 5-, or even 8-in.) scintillation crystal. Because the scintillation crystal was also the first means of detecting radiation, the scintillation detection process could be considered the beginning of scanning.

Scintillation Detection

Around the turn of the century, Sir William Crookes made a brass tube a little larger than the eyepiece of a microscope. One end was sealed and coated with zinc sulfide. Inside, in front of the coated end, he mounted a needle on a small thumbscrew to change the distance of the needle from the zinc sulfide screen. The needlepoint was dipped in a radium solution. A lens similar to a microscope eyepiece enclosed the other end. In looking through the eyepiece at the now completely dark screen, as the radium needle was brought closer to the screen, one could see that the dark background was dotted with bright flashes of light, which multiplied as the radium approached the screen. This first gamma-scintillation counting device was invented to look at scintillations from alpha particles from a radium source. More than a generation later, when the scintillation idea was being applied to medical gamma counting, the radium originally used was no longer acceptable for medical practice in unsealed sources

Crookes published the details of his "Spintharicope" (from the Greek words for "spark viewer") in 1903. Within 5 years, Rutherford and Geiger described

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the already "...well-known property of producing scintillations in a preparation of phosphorus and zinc sulfide..."[1] They pointed out that it wasn't difficult to count the number of scintillations, since they were visible light flashes. But they had no confidence that the number of alpha particles was the same as the number of disintegrations. Hence, they described a second—electrical—method. It was the forerunner of the Geiger-Mueller tube, developed around the principle of an electron avalanche measurement worked out by Townsend (an old Cavendish Laboratory schoolmate of Rutherford) at least 7 years before. This was the same principle that was to be used so effectively for beta counting a generation later.

Rutherford and Geiger had serious objections to the theoretical validity of the method unless one knew the decay scheme (remember that the profusion of artificial radioisotopes is still almost a third of a century in the future). In spite of the objections to scintillation counting, the Townsend–Rutherford–Geiger electrical method was almost abandoned for about 20 years in favor of the scintillation method.

What Field of Vision Do Physicians Want?

Still another confusion was related to the collimator problem—the field of vision desired. The modified Crookes vacuum tubes, first used to produce radiation, sprayed X rays in every direction. As early as 1896 a shield was put around the tube except for a hole that made a "beam" of radiation. Only low-energy X rays were being generated, and even a thick piece of glass, especially if it had some lead in it, was good radiation shielding. A few years later, Friedrich Dessauer (one of Germany's first radiation physicists) wrote his first article on the theory of the Röntgen apparatus.[2] He hints that there is more to beam direction than just the mechanical aiming of the hole in the shield. There is also such a thing as a depth dose along the central axis (later, on other axes). Depth dose was crudely measurable but not, at that time, calculable.

By the end of the first World War, the more sophisticated radiotherapists were using Dessauer charts: two-dimensional representations of lines of equal dose as a beam of radiation passes through a body, usually a water "phantom." Shortly after World War I, a group of American radiotherapists went to Germany to see what was going on in radiology. A very active school in radiation physics was centered around the great German radiation physicist Walter Friedrich; one of his students was Otto Glasser.

In spite of his friendship with Dessauer, Glasser was not satisfied with the eponym and gave the name "isodose" charts to these Dessauer charts. The word rapidly took hold. In 1923, when Glasser moved from Germany to the Cleveland Clinic, an article was published in *Radiology* on Dessauer charts; another referred to isodose charts. Glasser was using isodose measurements not only for a beam of radiation but also for the distribution of dose around a radium needle. These isodose lines were physical forerunners of the scanning idea.

Isodose Curves Are a Method for Measuring Field of Vision

Around 1928, an English radiation physicist, W. V. Mayneord, appeared on the scene and eventually changed the entire field.[3] He described isodose curves as capable of being modified by a variety of conditions, the most important being the "effective wavelength." Up to that time (and for a long time thereafter), energy of radiation in medical fields was a matter of crude half-value layers. The idea of specific X-ray energies was developed around Bragg's 1912 crystal spectrometer, but it was difficult physical instrumentation. The idea of *half-value layer*, though a biased averaging process, was much handier for medical work. Actual medical spectrometry on a clinical scale did not begin until the crystal spectrometer was developed, largely by P. R. Bell's group in Oak Ridge, during the 1950's. But the sodium iodide crystal had to wait for a photomultiplier tube, which in turn had to wait for a photoelectric tube and for various counting circuits which weren't really available until after the second World War.

During the 1930's, there were extensive studies of isodose curves by many American, English, and German radiotherapists and radiophysicists. There was a profusion of adaptations of the isodose curve idea to almost every problem in radiotherapy. In 1937, Mayneord, in discussing "The Significance of the Roentgen," while upholding the roentgen as a physical unit, pointed out that the real unit of dose is a problem of energy absorption in the tissues and that different tissues have different absorptions.[4] This was the beginning of a new unit of radiation dose, but it was not used until another English radiophysicist, F. W. Spiers, demonstrated the enormous effect, on dose, of differential tissue absorption. The essential point in the story of scanning is that Spiers' work was begun in conjunction with a study of materials used in measuring depth dose, and finally ended with the

recognition of a fundamental fault, not only in depth-dose measurements but also in the unit of measurement itself.[5]

While this demonstration was being made, Mayneord carried the isodose curve idea through two-dimensional calculations of what the dose might be and extended the curves to three-dimensional surfaces. Isodose surfaces are what a source of radiation produces; field of vision is what a radiation detector sees.

During and shortly after World War II, clinical development largely lay fallow. When it reopened in 1949, the British radiotherapists held a conference directed primarily toward the problem of radiologic localization. The newer ideas went back to Mayneord's concept of isodose surfaces.[6]

How Radioisotope Scanning Began

As a natural extension of the "point measurements" for determining the dosimetry field around a radium needle or in a beam of X rays, the English developed a system, before 1950, of making a series of "point measurements" over the thyroid gland. By the mid-1950's most of the larger English hospitals had templates that would fit over the neck and were marked off in 1-cm squares. Points at equal count levels were joined to form "isodose lines." These lines made a "picture" of the distribution of the radioiodine in the thyroid gland, but they weren't doses (and no one thought they were); they were the responses of a detector. A better system with crystal detectors was being developed in the United States, but the English physicians could not afford crystals. Benedict Cassen's mechanically moving "beam" of detection is a logical outgrowth of this multiplicity of regularly spaced point measurements.

While the isoresponse-point measurement system was developing in England, even before 1948, the Moore group at the University of Minnesota was developing a technic using diiodofluorescein labeled with 8-day ^{131}I to localize brain tumors.[7] Radiophosphorus uptakes had been in use for a number of years for this purpose, using remarkably small Geiger tubes in needles, which were inserted directly into the brain. But the Moore group wanted external measurements.

At first they did an iodine uptake measurement over one point on the skull. Then they went to multiple symmetrical counting positions and, finally, when the early scanners became available, to a scanning procedure. This is not an uncommon pattern of development. The same thing happened with localizing liver defects, and it has just happened in placentography.

Growth of Scanning Idea

The growth of a medical idea is reflected in the volume of the literature. For me to state how many articles have been published in the medical literature on scanning depends, of course, on how I am going to define the word "scanning." This is especially true in the pre-1950 literature. If I twist the word to include point counting, external uptake counting, and the developmental articles so essential to the introduction of the idea, then the literature on scanning before 1950 is very large. However, if I restrict the word very arbitrarily to what I think is essentially what we now know as scanning, then before 1948 there were only 18 articles. In 1948–1949 there were 35, including the article by Cassen that I am (again rather arbitrarily) listing as the original significant reference. During the first half of the 1950's, there were 158 articles, of which about 10 marked the beginnings of almost everything that is now being done. In the latter half of the 1950's, there were only 173 articles on scanning, of which very few marked new ideas.

Beginning in 1960 with about 50 articles per year published, the field explodes to over 257 in 1964. However, during this period there are fewer and fewer articles on essential ideas and fewer on instrumentation. There is an ever-increasing concern with new labeled compounds and with the less familiar radioisotopes. Most of the articles in the 1960's are on experiences in clinical application of the scanning idea. Many are repetitions.

Very generally stated, the 1940's are mostly a period of ^{32}P beta counting, with a rapid shift toward ^{131}I (gamma emitter) uptakes. The 1950's start out with a shift to mechanical scans, a very rapid shift from Geiger-Mueller to crystal scintillation detection, and then an even more important shift to scintillation spectrometry. Pharmacologically, there is a shift from raw, inorganic radioisotopes to ever more specific labeled compounds.

Clinically, scanning starts with brain-tumor localizations and finds its major development in thyroid-gland visualization; then extends to spinal cord, liver, spleen visualization, heart, gallbladder, bone metastases, bone marrow, and placenta; and, by 1960, to whole-body scanning (this is distinct from whole-body counting, an entirely different development). The 1950's saw many trials on patients. During the 1960's, the trials on patients became long series, and finally, by

1964, almost routine reports, of clinical experience. On July 1, 1966, my scanning bibliography contained 1201 entries.

In January 1958, the first textbook on radioisotope scanning was published.[8] In October 1958, the first symposium on scanning was held. By 1964, there were 14 major book-length publications in the field—usually proceedings of symposiums. By 1966, American (but not Japanese, English, or German) publications were confusing the words "nuclear medicine" with "scanning."

Instrumentation for the scanning procedure started out with a commercial development in 1950. By 1966, 14 major companies were producing equipment in at least seven different countries (including Japan, which I mention specifically because it is so often forgotten and is so important in new developments).

Where Do We Go From Here?

Scanning radioisotopes was a concept in 1950; it was almost a clinical necessity by 1965. Five new developments appeared (in the late 1950's) that foreshadowed a complete change within the next decade or so:

1. Positron scanning, an early 1950 idea held back by the unavailability of positron emitters, is beginning to reassert its theoretical superiority. The annihilation radiation from positron emitters is no longer the essential item, but the opposed-beam technic that grew out of it is.

2. A double collimator "B filter" system is developing. This double system (an American–Japanese development) uses two matched counters: one measures everything in the field, the other shields the directly collimated umbra of the beam. By subtraction, the unwanted penumbra of the field of vision is annulled.

3. A rescanning system (an Oak Ridge development) fits any idea in spectrometry, recording, or collimation design. There are a tremendous number of factors of discrimination in any scan pattern. Should counts be averaged over a short or a long period? Should every count be looked at? What kind of marking? Speed? Background? Color? Much of this physical discrimination can be done only by the interpreting physician who doesn't know what he wants to see until after he has seen it. The inconsistency is resolved by measuring all the information on one scan (resulting in an uninterpretable picture). By rescanning the original scan, the clinician can select which factors of discrimination he thinks are best for his purpose.

4. Color display is a Japanese idea. The recording system was started with a simple dot recording. Then there was a photographic improvement. At first, color recordings were nothing more than pretty pictures and were commonly called "Christmas-card scans." However, clinical interpretation is the only important end product of any scan. The fact that a good physicist can interpret a black-and-white scan just as easily as a color scan does not vitiate the idea that even an untrained clinician can see color localizations far more easily than black-on-white dots. Color recording is important.

5. The J. S. Laughlin group in New York is approaching the whole body with a "coarse scan" idea that is 50% whole-body count, 50% whole-body scan, and 50% whole-body uptake. This totals 150% and proves that the whole is better than any of its parts. I don't like their solution, but I think it's the direction of the future.

I have neglected a thousand things in this brief historical assessment. I am talking primarily about the instrumentation built up around the subject of scanning. However, whether the instrumentation changes in the next few years or not, the simple release of a couple of the new radioisotope "cow systems" will allow increased dose and decreased scanning time, even with the present machinery of scanning.[9] The discovery of a couple of new compounds would have an importance far exceeding anything that can be done in modifying instruments.

There Will Be More Than One Good Scanning System

But even within the strict field of instrumentation, a number of developments are occurring that are not really new but are extensions of trends already occurring. For example, the future is undoubtedly going to see the use of larger crystals in order to get greater sensitivity of detection for achieving more rapid procedures.

Collimator design has stagnated since the mid-1950's. But, with the development of greater detection sensitivity, there is going to be more freedom in achieving a desired field of vision. The use of multiple-crystal systems along with greater freedom in collimator design is going to improve the very serious limitations in spatial resolution. When the anatomic pathologist speaks of "spatial resolution," he talks in terms of micron measurements. The diagnostic radiologist talks in terms of fractions of a millimeter. The scanning specialist must talk in terms of centimeters.

This position will not be tenable for very long. But probably most significant of all is the simple growing realization by many physicians that there is more than one system of scanning.

As scanning becomes more common, it will lose some of its present curse of being a publicity gimmick. As more physicians who do not do scanning talk to more scanning technicians and learn to demand a little more awareness of clinical problems, much better reporting systems will be developed. There is currently a lot of discussion by manufacturers of equipment concerning very fancy recording and display computers. This is all to the good, but few people seem to be worried about the much more fundamental problem of reporting the results of a scan. Just how do you tell a practicing physician that "this picture looks like metastatic carcinoma in the liver, but maybe it ain't?"

The whole problem of scanning time, which is considerable even with the so-called fast systems, is beginning to receive some consideration. But it is still difficult to convince an instrumentation engineer who has never seen a patient that the essential element in the time span is not the microsecond data transfer in a computer, but the half-second distortion on a record when the patient scratches his nose.

Medical radioisotope scanning is finally beginning to achieve the popularity that 15 years ago we were predicting would occur momentarily. Back in the days just after World War II, the radioisotope had achieved an importance "second only to the microscope." The radioisotope was "going to lay bare the innermost secrets of biochemical metabolism." Only a full radiobiological assay of the distribution of many labeled compounds could hope to approach (not achieve) such an ideal. Radioisotope scanning is not, and never will be, a full assay, nor is it likely to reach the ideal. (MG)

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Section

VII

Isotopes and Radiation Technology

Isotopes Around the World

Russian Irradiation Facility Development

Among recent interesting Russian isotope developments are two irradiation facilities: the first USSR industrial radiochemical plant, which uses ^{60}Co in the manufacture of sulfonate detergents, and a commercially produced mobile irradiator, which uses ^{137}Cs for irradiating seeds prior to sowing. Both facilities were described in the November 1967 issue of *Atomnaya Energiya.*[1,2]

Radiochemical Plant

The first industrial radiation-chemical facility in the USSR is in continuous operation, having been started up in January 1967. Using a method developed at the All-Union Scientific Research Institute of Radiation Technology, the plant produces the raw material for the synthetic sulfonate detergents from paraffin hydrocarbons, SO_2, and Cl_2. The gamma radiation from ^{60}Co is used to initiate the reaction. The facility is compact, its individual units are dependable, and long maintenance-free periods are possible. Production rates as high as 500 kg/hr are possible.

The reaction vessel is cylindrical and was installed directly in an existing chemical plant without construction of special canyons. It is surrounded by a cast-iron shield of thickness sufficient to reduce the radiation dose to personnel at the outer shield surface to appreciably below permissible limits. This provision for biological shielding decreases capital costs considerably over what they would be if canyons had to be constructed.

The linear source is a 2- to 4-m-long assembly of spherical ^{60}Co units arranged along the axis of the reaction vessel. The radioactive spheres may be interspersed with nonactive spheres.

The reaction ingredients are introduced countercurrently, and both reagent introduction and product withdrawal may be either continuous or in batches. Homogeneity of the reaction mixture in the radial direction is achieved by using bubbles of gaseous Cl_2 and SO_2.

Cost estimates indicate annual operating costs of 200 thousand rubles ($220 thousand).

Seed-Irradiation Facility

Russian scientists have been especially active in the area of irradiating seeds to produce desirable mutations or hasten germination, and the results of their work (see Refs. 3 to 6, for example), extending over a period of years, have indicated that definite benefits may often be expected from low-dose irradiation (Table 1). In order to gain industrial acceptance of the technique—in particular, for cotton, potatoes, grain, and vegetables—the USSR isotope production group, *Izotop,* is now manufacturing[7] a mobile irradiation facility which can be operated in the laboratory from a 220-volt line current but which has batteries for field use. The equipment, which has been under development for some time,[8,9] weighs 3000 kg and, along with a motor, is mounted on a wheeled base.

The doubly encapsulated 2100-curie radiation source has a cylindrical arrangement to ensure dose homogeneity. The 210-mm-diameter cylinder is assembled from eight units, each of which is 14 mm in diameter by 242 mm long. The dose rate in the irradiation chamber is 600 r/min. Shielding is sufficient to ensure that operating personnel will not receive doses greater than permissible. Operating convenience is provided by the two-chamber construction, each

Table 1 Results of 5 Years of Research on Seed Irradiation

Crop	Radiation dose, r	Effect		
		Description	Laboratory conditions	Production conditions
Radish	1000	Shortened ripening period	5–7 days	4-6 days
		Increased fertility	20-30%	11-20% (for two seasons)
Carrot	2500	Increased fertility	24-30%	16-30%
		Increased carotin content	6-12%	6-12% (for three seasons)
Corn	500	More abundant foliage and sweeter corncobs	15%	12-20%
Cucumber	300	Increased fertility	15-20%	10-15%
Lupin and vetch	50–150	Increased fertility	15-20%	10-15%
Rye	1000	Increased fertility	20%	–
Turnip	500	Increased fertility	37%	–

with a volume of 1 liter. This permits irradiation of materials in one chamber while the other is being loaded.

Gamma-radiation doses recommended[4] for seed irradiation are: for peas, 300 r; cucumber, 300 r; turnips, 500 r; corn, 500 r; rye, radish, and tomato, 1000 r; cabbage, 2000 r; and carrot, 2500 r.

(Martha Gerrard)

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Rapid Curing of Coatings by Electron Bombardment

Twenty-three people met[1] at Saclay, France, on Nov. 3, 1967, to discuss the use of electron irradiation to induce polymerization, cross-linking, or grafting—and thus hardening—of liquid or viscous monomers deposited on a surface. The participants were representatives of the paint industry, accelerator manufacturers, and the Commissariat à l'Énergie Atomique (CEA).

Theoretical bases of the reactions were discussed, after which attention was given to practical problems. For example, for the two thicknesses of paint in

common use, 20 to 50 μm, the use of 300- to 500-kev electrons is considered economically feasible. The procedure results in a saving of time and perhaps of raw material, but it cannot yet be used for very irregular surfaces such as automobile bodies.

Work done in England and the USA was cited, together with descriptions of the Texas Nuclear and Radiation Dynamics equipment, the Ford Motor Co. patented process, and the Boise-Cascade installation.[2-4] A facility being installed by Radiation Dynamics at Stuttgart was mentioned. The French have two suitable accelerators: (1) the CIRCE, installed at Corbeirele Essonnes, whose beam can be varied from 1 to 10 kw; and (2) the SAMES equipment, which is to be constructed under a CEA license and put on the market; its characteristics are a 3-kw beam and capability of irradiating a 1- by 0.5-m surface.

The complete proceedings of the meeting, including discussion, will be published in the journal *Peinture, Pigments et Vernis.*

In England a prototype electron-beam finishing line is in operation at a paint factory, and a commercial plant is being planned by a furniture manufacturer.[5,6]

(Martha Gerrard)

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Section VIII

Isotopes and Radiation Technology

Miscellaneous

Availability of Isotopes and Services

IAEA Radiation Standards

In 1968, as part of its program of distributing standardized radioactive samples for use in checking measuring equipment, the IAEA will supply, to laboratories using radioisotopes, calibrated solutions, solid gamma sources, sets of four activation detectors for determining fast-neutron spectrums, and sulfur pellets containing ^{32}P for measuring fast-neutron flux densities and fluences. Information on the program may be obtained from the International Atomic Energy Agency, Kärntner Ring 11, A-1010, Vienna, Austria (reference LAB/242-7).

ORNL Stable-Isotope Inventory

Recent additions to the ORNL enriched-stable-isotope inventory include the following:

Isotope	Purity, %	Amount available, g
^{33}S	9.8	15.7
^{36}S	0.2	2.9
^{36}S	1.5	0.4
^{87}Sr	82.6	13.3
^{97}Mo	94.2	0.5
^{150}Nd	95 to 97	18.1
^{168}Yb	18 to 22	11.2
^{176}Yb	84.3	12.8
^{196}Hg	47.5	0.8
^{199}Hg	86.1	1.1

(PSB)

Nuclear Equipment To Enter Radiochemical Production Field

Nuclear Equipment Chemical Corp., of Farmingdale, N. Y., is planning to produce radiochemicals and radiopharmaceuticals, which the company formerly imported from France. They will use the test reactor and laboratory facilities of Industrial Reactor Laboratories, Plainsboro, N. J. They are also negotiating with Neutron Products, Inc., for purchase and installation of a 50-kilocurie ^{60}Co source.

Gamma Process Co. Reports Prosperity

For the year ending Jan. 31, 1968, Gamma Process Co. reports earnings of $41,086 on revenues of $337,749. This is a substantial change in the picture over FY 1967, which showed a $20,126 loss on revenues of $72,214. The company is planning installation of a radiation processing plant near Buffalo, N. Y.

Limited Amounts of ^{238}Pu To Be Sold

The Atomic Energy Commission is making available for sale, for the first time, 500 g of ^{238}Pu for use in private research and development. The material, which has an isotopic purity of 80%, will be sold from the AEC's Oak Ridge (Tennessee) National Laboratory at $1000 a gram, as the oxide.

The best known use of ^{238}Pu, which is produced by irradiation of ^{237}Np in a nuclear reactor, is as an isotopic heat source for small power units, e.g., those for use in satellites. Its long half-life, high alpha output, and low-energy gamma emission make ^{238}Pu useful in alpha and neutron sources for gaging, static elimination, and oil-well logging. It may also be useful for

medical applications such as heart pacemakers and as the power source for a mechanical heart pump.

Any person purchasing ^{238}Pu must have a Special Nuclear Material License issued by the U. S. Atomic Energy Commission. Requests to purchase research quantities of ^{238}Pu should be referred to Isotopes Sales Department, Oak Ridge National Laboratory, Post Office Box X, Oak Ridge, Tenn. 37830.

AEC Activities

Division of Isotopes Development-Sponsored Research and Development Work Summarized

"Division of Isotopes Development Research and Development Projects—1967" is now available as TID-24349 from CFSTI. The project summaries are considerably more detailed than the brief summaries furnished by the contractors to Science Information Exchange of the Smithsonian Institution [see *Isotopes and Radiation Technology*, 5(1): 63-82 (Fall 1967)]. This 290-page report summarizes not only the objectives but the important results and future plans for each of 118 research and development projects, both at Commission installations and with private contractors, sponsored by the Division. The projects are presented according to the seven DID program areas—Radioisotopes Production and Materials, Thermal Applications, Environmental and Ocean Science, Radiation Analysis and Control, Radiation Preservation of Foods, Process Radiation, and Radiation Engineering—and reflect the status of the projects as of November 1967. The project summaries were prepared by the contractors and edited by staff members of the Isotopes Information Center. (JEC)

Isotope Heater for Aircraft Guidance Systems

A heater fueled with ^{147}Pm, developed by the USAEC and the Air Force, has been delivered to the Air Force for testing its capability of keeping aircraft guidance systems near the proper operating temperature. The gyroscopes and accelerometers used as sensors in inertial navigation systems are not reliable until they reach a stable operating temperature, and some time may be required for this temperature to be reached, especially in cold areas. The ^{147}Pm heater would eliminate the warm-up period by keeping the entire unit near the sensor operating temperature. Operation of the heater depends on transfer of heat from the isotope capsule to the sensors by circulation of an ethylene glycol solution. When the sensor reaches the proper temperature, a valve shifts the fluid flow to an external heat exchanger to dissipate the heat. The technique is practical because these sensor systems are now small enough that less than 60 thermal watts will keep them at operating temperature. Safety of the heater fuel capsule, which is an approximate sphere of 2 in. diameter, has been demonstrated in sled-impact and airdrop tests at the AEC's Sandia Laboratory in Albuquerque, N. Mex. The measurement unit and thermal preconditioner were fabricated by the AC Electronics Division of General Motors Corp. in Milwaukee, and the heat source was fabricated by Pacific Northwest Laboratory at Richland, Wash., operated for AEC by Battelle Memorial Institute. The heater units will be assembled and ground tested in the laboratory at Wright-Patterson Air Force Base, Ohio, to determine operational characteristics for temperatures as low as $-65°F$. Flight tests will be made at Holloman Air Force Base, N. Mex.

Radioisotope Licensing Changes

The USAEC has proposed amending Part 150 of its radioisotope licensing regulations in an attempt to ensure uniform standards for regulating transfer of products containing small amounts of radioactive material. Currently, although all such products must conform to safety standards detailed in a specific license obtained from the AEC by the manufacturer, some of them may be used by persons without the requirement of a license. As the number of industry proposals for exempt products increases, it will become increasingly difficult to determine whether or not a product will be used by the general public. The proposed changes would give AEC the authority over the transfer of all these products instead of, as now, only over those intended for use by the general public. The amendments would apply to products manufactured in agreement states to which the AEC has transferred part of its licensing authority.

Prices for Two Radioisotopes Being Lowered

The Atomic Energy Commission is lowering the prices on ^{237}Np from $500 per gram to $225 per gram, effective Apr. 19, 1968, and on ^{241}Am from $1500 per gram to $1000 per gram, effective Jan. 1, 1969. The new prices result from the introduction of new, more efficient production processes and the processing of increased amounts of material.

Neptunium-237, which has one of the longest half-lives of any of the transuranium elements (more than a million years), is useful in the study of the chemical and physical properties of heavy, man-made elements. Americium-241 is important in the production of neutron sources, which are widely used in research, medicine, and industry. Both nuclides are essential intermediate products in the production of ^{242}Cm and ^{238}Pu, which hold promise as isotopic heat sources for small auxiliary electric-power units. Requests for further details and information on purchase of the two radioisotopes should be sent to Isotopes Sales Department, Oak Ridge National Laboratory, Post Office Box X, Oak Ridge, Tenn. 37830.

Radioisotope License Guides

"How To Get a License To Use Radioisotopes" is a 35-page booklet that gives complete information on state regulations, AEC safety standards and regulations, conditions under which licenses are required, and how to go about obtaining a license to use radioisotopes in amounts ranging from the microcurie quantities used in medical diagnosis to the megacurie quantities used in isotope-powered generators. Instruction sheets and sample applications are included in an appendix.

"Industrial Radiography" is a 71-page guide on radiation safety considerations in the preparation of license applications for authorization to use sealed sources in radiography. Information is included on radiography personnel qualifications, operating and emergency procedures in the use of sources, inspection and other internal control systems, organization of the radiography program, leak testing, how to complete an application, and radiographic operations in agreement states. Various details are given in appendixes.

Both booklets are published by the Division of Materials Licensing of the USAEC and are sold by the

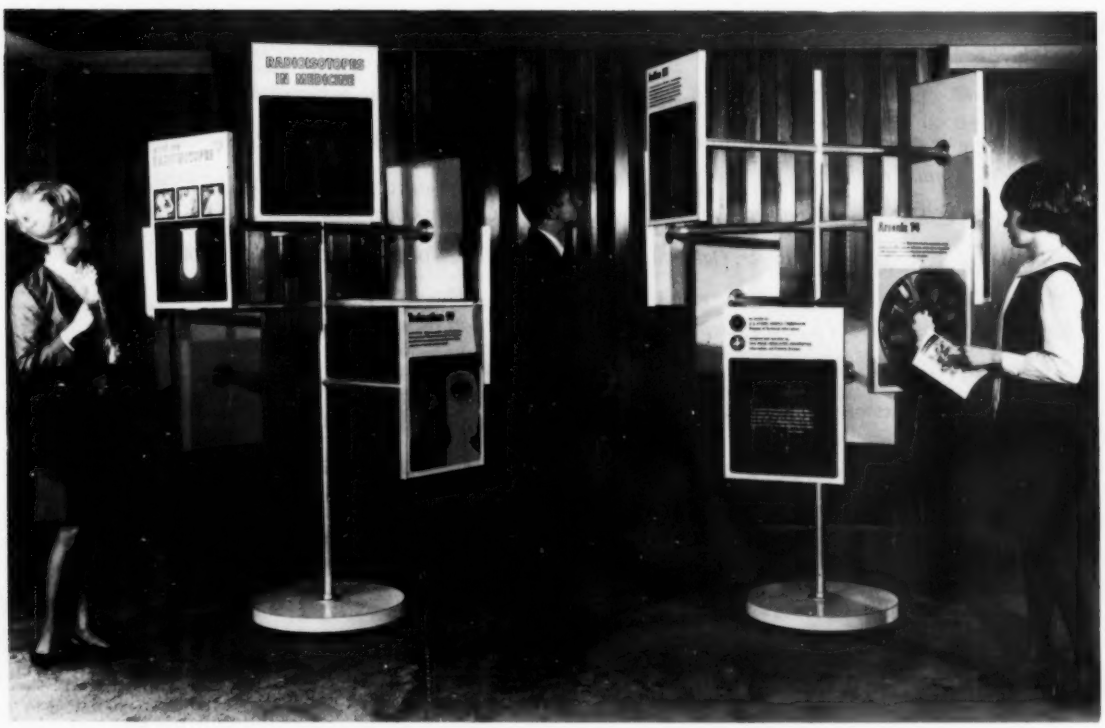

Fig. 1 "Radioisotopes in Medicine" exhibit.

Superintendent of Documents, U. S. Government Printing Office, Washington, D. C. 20402. "How To Get a License" costs 30 cents and the radiography guide, 50 cents.

"Radioisotopes in Medicine" Exhibit

A new exhibit, describing the varied and expanding medical uses of radioisotopes, has been designed and constructed by Oak Ridge Associated Universities for the U. S. Atomic Energy Commission. The exhibit, "Radioisotopes in Medicine," is composed of two metal "trees," each with eight "branches" (Fig. 1). On each branch is a panel describing applications of radioactive elements in the diagnosis and treatment of diseases, e.g., ^{137}Cs and ^{60}Co for treatment of deep-seated tumors, ^{131}I for diagnosis and treatment of thyroid disorders, ^{87m}Sr for studying bone disorders, ^{90}Y for pituitary-gland ablation, and ^{99m}Tc for detecting brain tumors. The exhibit can be assembled and disassembled in a remarkably short time, has no electric-power requirements, and can be set up in a relatively small display area. It is particularly suited for showing in libraries, schools, and hotel or office-building lobbies and is available without charge to qualified sponsors. Information about scheduling showings of "Radioisotopes in Medicine," as well as other exhibits, may be obtained from the Information and Exhibits Division, Oak Ridge Associated Universities, P. O. Box 117, Oak Ridge, Tenn. 37830.

Program for Packaging and Transportation Meeting Announced

The program for the Second International Symposium on Packaging and Transportation of Radioactive Materials, cosponsored by Union Carbide Corporation and the U. S. Atomic Energy Commission and to be held in Gatlinburg, Tenn., Oct. 14–18, 1968, has been announced:

Monday Afternoon, October 14

Welcome by L. B. Shappert, Symposium Chairman, Oak Ridge National Laboratory, Union Carbide Corporation, Oak Ridge, Tenn.

Welcome by C. E. Larson, President, Nuclear Division, Union Carbide Corporation, Oak Ridge, Tenn.

Welcome by S. R. Sapirie, Manager, Oak Ridge Operations Office, United States Atomic Energy Commission, Oak Ridge, Tenn.

Keynote Address, Clifford K. Beck, Deputy Director of Regulation, Washington, D. C.

SESSION I. Transportation Regulations and Experience in Their Implementation

Chairmen: J. A. Lenhard, United States Atomic Energy Commission, Oak Ridge Operations Office, Oak Ridge, Tenn.

L. R. Rogers, United States Atomic Energy Commission, Washington, D. C.

The Competent Authority—How Is He Coping? *E. J. Wilson and A. H. Partridge, Ministry of Transport, London, England.* Discussion on problems the competent authority is meeting in putting IAEA transport regulations into effect; explanation of NAIR (National Arrangements for Incidents Involving Radioactivity) scheme.

The New Regulations and Their Impact on the Transportation of Radioactive Materials. *Robert W. Blackburn, Atomic Energy Control Board of Canada, Ottawa, Canada.* Review of the effects of the new regulations on the general public, packaging designers, shippers, carriers, transportation facility operators, and regulatory authorities.

Main Problems Arising from the Application of IAEA Recommendations. *Y. Sousselier and J. Bouillet, Commissariat à l'Énergie Atomique, Paris, France.* Presentation of practical difficulties (administrative, theoretical, and technical) which have been met in domestic and international operations.

Present and Future Role of the Atomic Energy Commission in the Regulation of Radioactive Material Transport. *J. A. McBride, United States Atomic Energy Commission, Washington, D. C.* Discussion of the experience acquired in operations of AEC and DOT under the Memorandum of Understanding and the projected future role of the AEC in respect to approval of packaging for fissile and large quantities of radioactive material.

Implementation of the New AEC Transport Regulations—10 CFR 71. *Donald A. Nussbaumer and Charles E. MacDonald, United States Atomic Energy Commission, Washington, D. C.* Discussion of experience to date in implementing the new AEC transport regulations; possible changes in the regulations are identified and discussed.

Review of Implementation of Transport Regulations for Criticality Control. *G. H. Bidinger and R. H. Odegaarden, United States Atomic Energy Commission, Washington, D. C.* Review of the difficulties that licensees have encountered in satisfying the safety standards of Part 71.

The Coast Guard and Maritime Radioactive Materials Traffic. *Terry R. Grant, United States Coast Guard, Washington, D. C.* Discussion of problems encountered in transporting radioactive materials by merchant ships and the special provisions required by U. S. Coast Guard Regulations.

Administration of Radioactive Materials Transport—Oak Ridge Operations Office. *William A. Pryor and Thomas H. Hardin, Oak Ridge Operations Office, United States Atomic Energy Commission, Oak Ridge, Tenn.* Discussion of ORO safety control and traffic management aspects in shipping radioactive materials within rules of AEC, DOT, and IAEA.

The High Cost of Red Tape. *William A. Brobst, United States Department of Transportation, Washington, D. C.* Discussion of difficulties encountered by atomic energy industry in arranging for transportation of radioactive materials and the question of what we can do about the high cost of paper work requirements.

IAEA Regulations for the Safe Transport of Radioactive Materials. *G. E. Swindell, International Atomic Energy Agency, Vienna, Austria.* Discussion of IAEA plans for revision and long-term development of its transport regulations.

SESSION II. Transportation Experience and Problems

Chairmen: Duane R. Swindle, United States Atomic Energy Commission, Washington, D. C.

Robert A. Kaye, United States Atomic Energy Commission, Washington, D. C.

Tuesday Morning, October 15

REA Express Problems in the Transportation and Handling of Shipments of Radioactive Materials. *J. J. Toughey, Quality Control Services, REA Express, New York, N. Y.* Discussion of REA Express problems in handling shipments of radioactive materials with emphasis on packing and marking of containers.

Operating Experience in Radioactive Shipments—NRTS. *J. P. Byrom and A. A. Anselmo, Idaho Nuclear Corporation, Idaho Falls, Idaho.* Review of experience in shipping radioactive material from National Reactor Testing Station (NRTS) with emphasis on radiation, contamination, and tiedown problems.

Some Legal Problems in the Transportation of Radioactive Materials. *Edward G. Howard, General Attorney, Association of American Railroads, Washington, D. C.* Discussion of proposed amendments to the Price–Anderson Act.

A Fuel Receiver's Experience. *R. F. Rogers and J. F. Forstner, E. I. du Pont de Nemours and Co., Savannah River Plant, Aiken, S. C.* Discussion of experience in fuel receipt and storage and how the receiver's needs affect the shipper and reactor operator.

An Examination of Radium Losses During Transportation. *Gail D. Schmidt, National Center for Radiological Health, Rockville, Md.* Discussion of circumstances surrounding recent losses of radium sources during shipment.

Progress Report—AEC Headquarters, Division of Construction's Shipping Container Program. *J. A. Sisler, United States Atomic Energy Commission, Washington, D. C.* Review of container program including utility review procedure, specification containers, and container inventory with discussion of problems encountered in coordinating the program.

The Accident Experience of the USAEC in the Shipment of Radioactive Material. *D. E. Patterson, United States Atomic Energy Commission, Washington, D. C.* Review covering ~20 years' accident experience of USAEC in transporting radioactive materials and analysis of factors which have influenced this experience.

The Shipment of Spent Fuel from Power Reactors in the 1970's. *C. W. Smith, General Electric Company, Santa Clara, Calif.* Discussion of the shipment of the steadily increasing quantities of irradiated power reactor fuels involving probable trends in fuels, containers, transport environment, and administrative factors.

Pricing, Purchasing, and Managing Transportation of Radioactive Materials. *Charles F. Goodner, Richland Operations Office, United States Atomic Energy Commission, Richland, Wash.* Discussion of historical background of freight rates now in effect on radioactive materials, experiences encountered in negotiating rates on certain large movements, and some suggested do's and don'ts for both carriers and shippers concerning areas to avoid in making rates, selecting a carrier, and in physical handling of this order of traffic.

Unique Fast Reactor Fuel Characteristics Affecting Future Shipping Container Designs. *D. E. Bloomfield and C. L. Brown, Battelle Memorial Institute, Pacific Northwest Laboratory, Richland, Wash.* Discussion of unique problems in the design of casks and containers for transporting fast reactor irradiated and nonirradiated fuels.

SESSION III. Standards for Container Construction

Chairmen: John W. Langhaar, E. I. du Pont de Nemours and Co., Inc., Atomic Energy Division, Wilmington, Del.

William F. Black, American Association of Railroads, New York, N. Y.

Tuesday Afternoon, October 15

Brittle Fracture Problems in Relation to Thick-Walled Packagings for Radioactive Materials. *E. J. Wilson and A. H. Partridge, Ministry of Transport, London, England.* Discussion on suitability of certain types of packaging for shipments at ambient temperatures in lower end of temperature range.

Analytical and Empirical Methods in Structural Analysis of Shipping Casks. *J. H. Evans, H. A. Nelms, and W. C. Stoddart, Oak Ridge National Laboratory, Union Carbide Corporation, Oak Ridge, Tenn.* Description of test program and apparatus used in developing engineering information for evaluating shipping casks: derivation of empirical equations relating puncture energy to outer-shell thickness and ultimate tensile strength; test procedures for determining material properties; acceptability of weld joint design, weld quality, and postweld heat treatment under impact loading; discussion of validity of energy methods for the analysis of dynamic loading.

Development of the AEC Cask Criteria. *L. B. Shappert, Oak Ridge National Laboratory, Union Carbide Corporation, Oak Ridge, Tenn.* Summary of information that has been developed for cask criteria. The information, developed for R.D.T. primarily to determine if a cask meets the performance standards of the regulations, covers such topics as cask structural integrity, heat transfer, criticality, shielding, materials of construction, and fabrication.

Materials of Construction, Fabrication Controls, and Tests for Radioactive Materials Shipping Containers. *J. R. McGuffey, Oak Ridge National Laboratory, Union Carbide Corporation, Oak Ridge, Tenn.* Discussion of recommended materials, shop practices, and tests and inspection selected as part of the Shipping Container Criteria which is being prepared by ORNL for the AEC.

The Status of Standards for Tiedown. *John W. Langhaar, E. I. du Pont de Nemours and Co., Inc., Atomic Energy Division, Wilmington, Del.* Discussion of problems involved in developing a standard for cask tiedown systems.

Combined presentation of three papers published individually in the Proceedings: The Use of Thick Steel Wrappers for Large Lead Filled Flasks Which Will Obviate the Need for Special Thermal Shields, Reusable Capsules to IAEA Standards, The Use of Massive Foamed Polystyrene for Type A and Its Use with Other Materials for Type B Packaging. *Frank E. Dixon, United Kingdom Atomic Energy Authority, Harwell, England.* Discussion on designing and testing new containers and packaging components (e.g., thick steel wrapper plate, foamed polystyrene transport capsules).

SESSION IV. Fire and Impact Testing of Containers

Chairmen: C. L. Brown, Battelle Memorial Institute, Pacific Northwest Laboratory, Richland, Wash.

Barry D. Devine, Argonne National Laboratory, Argonne, Ill.

Wednesday Morning, October 16

The Problem of Demonstrating Resistance to Fire. *John W. Langhaar, E. I. du Pont de Nemours and Co., Inc., Atomic Energy Division, Wilmington, Del.* Discussion of problems in determining fire resistance in large shielded containers and efforts being made to more accurately predict temperature conditions and effects.

Assessment of the Thermal Behavior of Irradiated Fuel Casks. *K.-H. Veith, Bundesanstalt für Materialprüfung, Berlin, Germany.* Presentation of an analytical method which allows fire resistance of irradiated fuel casks to be assessed by computer programming.

Evaluation of a Wood Fire-and-Impact Shield. *R. J. Lauer and R. D. Seagren, Oak Ridge National Laboratory, Union Carbide Corporation, Oak Ridge, Tenn.* Description of a series of tests made to evaluate a wooden fire-and-impact shield after a period of use to determine its effectiveness.

Design and Testing of Curium Shipping Containers and Cask. *C. A. Wilkins, R. D. Kelsch, F. D. R. King, D. H. Stoddard, J. P. Faraci, and J. W. Langhaar, E. I. du Pont de Nemours and Co., Inc., Savannah River Laboratory, Aiken, S. C.* Description of containers and cask for shipping 500-g lots of curium and the scale-model testing program used to validate the design.

Drop and Fire Tests of Packaging in Sweden. *Roland Blomquist and Gunnar Jonasson, Aktiebolaget Atomenergi, Studsvik, Sweden.* Description of research testing and prototype testing performed at the Research Establishment of AB Atomenergi in Sweden.

Some Studies of Structural Response of Casks to Impact. *H. G. Clarke, Jr., The Franklin Institute Research Laboratories, Philadelphia, Pa.* Summary of results obtained for a variety of impact tests with small-scale complete and partial containers with a procedure for applying data in cask design.

SESSION V. Container Research, Development, Testing, and Analysis—Part 1

Chairmen: Frederick J. Shon, United States Atomic Energy Commission, Washington, D. C.

David R. Smith, Los Alamos Scientific Laboratory, Los Alamos, N. Mex.

Thursday Morning, October 17

Class I and Class II Packaging Concept for Dry Low-Enrichment Uranium Oxide Powder. *Thomas Gutman, Westinghouse Electric Corporation, Cheswick, Pa.* Description of new packaging concept to permit shipment of large quantities of dry low-enriched uranium dioxide powder with minimal container handling and turn-around time.

Development and Design of Celotex-Insulated Containers for Shipping Fissile and Other Radioactive Materials. *F. E. Adcock, Dow Chemical Company, Golden, Colo.* Description of design concept and test program for steel drum/Celotex shipping containers used for transporting radioactive materials.

New End-Loading Shipping Container for Unirradiated Fuel Assemblies. *A. J. Mallett and C. E. Newlon, Union Carbide Corporation, Oak Ridge Gaseous Diffusion Plant, Oak Ridge, Tenn.* Description of new end-loading cylindrical shipping container of improved annular design for Fissile Class I packaging.

A Family of Special Nuclear Material Shipping Containers. *R. E. Kropp, United Nuclear Corporation, New Haven, Conn.* Description of old (relicensed) and new containers designed to incorporate or consider the requirements of the new transport regulations; development and testing are discussed.

Computer Analysis of the Criticality Safety of Shipping Containers. *E. C. Crume, Union Carbide Corporation, Y-12 Plant, Oak Ridge, Tenn.* Discussion of the application of reactor-theory-based computer programs in assessing criticality safety of shipping containers.

The Design and Testing of a Laminated Steel Plate Cask for the Transportation of Irradiated Fuel Bundles. *W. R. Taylor, Atomic Energy of Canada Limited, Chalk River, Canada.* Description of new laminated steel-plate cask and testing program for establishing validity of design.

Test and Theoretical Analysis of Fire Resistivity of Beta-Ray Thickness Gauge Containers. *Tadao Moriya, Fire Research Institute of Japan, Tokyo, Japan.* Description of tests and analytical procedures used in determining fire resistivity of beta-source thickness gauges.

Survey of Fissile Material Shipping Containers for Both National and International Shipments. *David P. Wood, Barry F. Estes, and Edmund L. Barraclough, Albuquerque Operations Office, United States Atomic Energy Commission, Albuquerque, N. Mex.* Review of fissile material shipping containers recently developed by Dow Chemical Company (Rocky Flats Plant), Los Alamos Scientific Laboratory, Sandia Corporation, and Mound Laboratory.

SESSION VI. Container Research, Development, Testing, and Analysis—Part 2

Chairmen: J. E. Gillham, Pan American World Airways, New York, N. Y.

Arthur R. Matheson, Gulf General Atomic Corporation, San Diego, Calif.

Thursday Afternoon, October 17

Demonstration Fuel Element Shipping Cask from Laminated Uranium Metal—Design and Fabrication. *D. H. Stitt and V. A. Smith, Union Carbide Corporation, Paducah Gaseous Diffusion Plant, Paducah, Ky.* Description of shipping cask using laminated depleted uranium metal for shielding material, including design considerations and technological improvements applied in fabrication.

Demonstration Fuel Element Shipping Cask from Laminated Uranium Metal–Testing Program. *C. B. Clifford, Union Carbide Corporation, Paducah Gaseous Diffusion Plant, Paducah, Ky.* Description of nondestructive and destructive testing program used with demonstration fuel element shipping cask made from laminated uranium metal.

Advantages and Economic Aspects of Laminated Uranium Shipping Casks. *C. W. Loveland, Union Carbide Corporation, Paducah Gaseous Diffusion Plant, Paducah, Ky.* Discussion of advantages associated with laminated uranium cask construction, cost factors developed, and commercial production aspects of the developed technology.

The Alpha, Beta, Gamma Transport Packagings. *M. Labrousse and H. Bernard, Commissariat a l'Energie Atomique, Paris, France.* Description of new casks designed particularly for transporting quantities of plutonium.

Design, Analysis, and Fabrication of Shielded Containers to Transport HNPF Fuel Elements. *Harold N. Miller, G. C. Gower, D. E. Beaderstadt, Chicago Operations Office, United States Atomic Energy Commission, Argonne, Ill.; J. E. Owens, Atomics International, Canoga Park, Calif.; and P. A. Halpine, United States Atomic Energy Commission, Washington, D. C.* Review of Hallam Nuclear Power Facility (HNPF) fuel removal operation with emphasis on development and design of shipping casks.

Structural Considerations in the Design of the HNPF Six-Element Irradiated Fuel Shipping Cask. *R. I. Jetter, Atomics International, Canoga Park, Calif.* Description of Hallam Nuclear Power Facility (HNPF) irradiated fuel shipping cask and two aspects of the structural design.

Design of Containers for Protecting Concrete Shipping Vaults Against "Hypothetical Accident Conditions." *S. G. Pearsall, S. J. Majeski, and L. Gemmell, Brookhaven National Laboratory, Upton, N. Y.* Description of nonreturnable reinforced concrete vaults designed for shipping radioactive waste to disposal areas and the test program used to evaluate such containers under the new regulations.

A Shipping Container for Heat Generating Nuclear Materials. *R. J. Bass, E. I. du Pont de Nemours and Co., Inc., Savannah River Plant, Aiken, S. C.* Description of three-part shipping assembly for transporting heat generating fissile materials which are gamma and neutron emitters with gross alpha contamination potential and the container testing procedures used.

Combined presentation of two papers published individually in the Proceedings: The Design of Thermal and Mechanical Shields for Lead Filled Containers up to Ten Tons in Weight with Emphasis on Those Emitting Heat, A New Standard Series of Containers for Loads up to 40,000 Curies of ^{60}Co or Equivalent. *Frank E. Dixon, United Kingdom Atomic Energy Authority, Harwell, England.* Description of standard series of containers for up to 40,000 curies of ^{60}Co and design of thermal and mechanical shields for lead filled containers, including heat transfer measurements of shields in use with 150,000 curies of cobalt.

SESSION VII. Panel Discussion on Model Testing

Thursday Evening, October 17

Panel Discussion

Chairman: Reuben W. Peterson, National Lead Company, Wilmington, Del.

Moderator: John W. Langhaar, E. I. du Pont de Nemours and Co., Inc., Atomic Energy Division, Wilmington, Del.

A panel composed of people with experience in model testing will discuss various aspects of model test program planning, model design, and evaluation of results. High-speed movies of cask impacts will be shown, and the applicability of test results to full-size casks will be evaluated. Open discussion will follow the initial presentation.

SESSION VIII. Container Research Development, Testing, and Analysis—Part 3

Chairmen: Nathaniel Welch, Federal Representative to Southern Interstate Nuclear Board, Atlanta, Ga.

Robert Kennedy, Department of the Army, Williamsburg, Va.

Friday Morning, October 18

Are the Type A Tests Realistic? *Frank E. Dixon, United Kingdom Atomic Energy Authority, Harwell, England.* Discussion of tests and results which tend to support thesis that some regulatory requirements are too restrictive.

The Release of Radioactive Material in the Event of the Destruction of a Full Type A Package. *S. Hagsgård, G. Jonasson, S. F. Johnson, and S. Henriksson, Aktiebolaget Atomenergi, Studsvik, Sweden.* Discussion of series of tests designed to justify IAEA restrictions on the amounts of friable solid radioactive material which may be transported in a Type A package.

A 30-Ton Cask for Shipment of PWR-BWR Fuel. *Armand Aupetit, Transnucleaire S.A., and Paul Blum, Society Lyonnaise de Plomberie Industrielle, Paris, France.* Discussion of reasons for choice of the 30-ton cask until the mid-70's, the problems involved, and some technical solutions.

Analysis of the Mechanical Response of a Cask to a Fire Environment. *K. D. Doshi, The Franklin Institute Research Laboratories, Philadelphia, Pa.* Discussion of procedures for evaluating the structural response of shipping containers under pressure loadings and at elevated temperatures.

Crash Studies of Highway Cargo Vehicles Carrying Typical Radioactive-Materials Containers. *R. A. Meldrum, Directorate of Transportation, Department of the Army, Williamsburg, Va.*

Engineering Considerations—Plutonium Nitrate Polyethylene Shipping Bottles. *D. A. Hoover and W. P. Ingalls, Atlantic Richfield Hanford Company, Richland, Wash.* Discussion of complex engineering problems associated with packaging, shipping, and unloading liquid fissile materials.

Summary, J. A. McBride, United States Atomic Energy Commission, Washington, D. C.

General

Marketing of Irradiated Onions in Canada

Fifty tons of irradiated onions {onions have already been cleared for food use in Canada [*Isotopes and Radiation Technology*, 5(1): 84 (Fall 1967)]} will be marketed through regular Canadian commercial outlets this spring. The onions are processed at the Atomic Energy of Canada Ltd. plant at St. Hilaire, Quebec, which uses ^{60}Co as the source of radiation. The plant was formerly operated by Newfield Products Ltd., which, before it went out of business, processed potatoes with radiation to prevent sprouting. Potatoes were the first irradiated vegetable to be marketed in Canada, and onions are the second.

Army's Ham Petition Not Acceptable to FDA

On April 19 the Food and Drug Administration (FDA) requested that the U. S. Army withdraw its petition for FDA approval of radiation-sterilized canned ham. Inadequate supporting data were cited as the reason for the action. The request is expected to have little effect on the AEC's low-dose irradiation of foods program; FDA has indicated approval of AEC protocols so far. The effect on the IRRADCO (Irradiated Foods, Inc.) pilot-plant meat-irradiator project has not yet been determined. (FEM)

Isotope Production in Poland and Argentina

According to a recent announcement in *Nucleonics Week*, the power of the Polish reactor at Swierk has been increased to 10 Mw(t), which should make Poland self-sufficient in the production of cobalt and iridium. On the other side of the Atlantic, the commissioning of the fourth Argentinian reactor, a 5-Mw(t) pool-type facility, may make Argentina an exporter of short-lived isotopes.

Irradiation Facility Design and Testing Services

Radiation Facilities, Inc. (63 Dell Glen Avenue, Lodi, N. J. 07644) has announced a complete service for planning, designing, fabricating, installing, checking out, and servicing of radiation processing facilities. The company has completed contracts with the AEC Division of Isotopes Development and other government agencies and has installed a number of small irradiators on a turn-key basis.

Japan–USA Cooperation in Ethylene Polymerization

In a cooperative project, Takasaki Laboratory of the Japan Atomic Energy Research Institute is study-

ing the radiation polymerization of ethylene at temperatures below the polyethylene melting point, while Brookhaven National Laboratory, in New York, will make similar studies at temperatures above the melting point. The information obtained is being exchanged between the two laboratories.

IAEA Issues Two Pamphlets in Safety Series

In late 1967 the International Atomic Energy Agency (IAEA) issued publications Nos. 9 and 23 in their Safety Series: "Basic Safety Standards for Radiation Protection (1967 Edition)" (STI/PUB/147) and "Radiation Protection Standards for Radioluminous Timepieces" (STI/PUB/167).

The Basic Safety Standards booklet is a revision of STI/PUB/26 and includes changes introduced on the basis of recommendations made by the International Commission on Radiological Protection and deals with (1) limitation of doses for exposure from controllable sources (i.e., maximum permissible doses for workers, dose limits for individual members of the public, and dose limits for the whole population); (2) radiation doses and intakes of radioactive materials through external and internal exposure (i.e., workers and individual members of the public); and (3) fundamental operational principles, describing general requirements as well as inspection and enforcement. Extensive tables of appropriate data are included, with some recommendations for partially unknown mixtures of radionuclides. (79 pages, available for $2 from National Agency for International Publications, Inc., 317 East 34th Street, New York, N. Y. 10016)

The Radioluminous Timepieces pamphlet is a new publication representing the recommendations of a group of experts (from Belgium, Czechoslovakia, France, Federal Republic of Germany, Japan, Switzerland, the United Kingdom, and the USA) set up by the IAEA, the European Nuclear Energy Agency (ENEA), and the Organization for Economic Cooperation and Development (OECD). Included are (1) the general and special requirements related to the radionuclides permitted (^{3}H, ^{147}Pm, ^{226}Ra) for use in luminous paint for dials and hands of timepieces; (2) control procedures for preparing and evaluating the timepieces; and (3) radiation protection considerations and determination of maximum permissible activities of the recommended radionuclides. (37 pages, available for $1 from the National Agency for International Publications, Inc., 317 East 34th Street, New York, N. Y. 10016) (PSB)

Nuclear Science Glossary Revised

The (U. S.) National Research Council and American Society of Mechanical Engineers nuclear science glossary originally published in 1957 has been revised. The revision, *USA Standard Glossary of Terms in Nuclear Science and Technology,* USAS N1.1-1967 (revision of N1.1-1957), was sponsored by the Atomic Industrial Forum, Inc., and is published by the United States of America Standards Institute, 10 East 40th Street, New York, N. Y., 10016. The glossary sells for $7.95.

New IAEA Radioisotope Projects

The IAEA has announced its cooperation—through supplying either equipment or technological experts—with university or government agencies of a number of countries in a variety of projects using radioisotopes. Radioisotope production will be initiated or expanded in Peru, Argentina, Venezuela, and the Philippines. In Indonesia, radioisotopes will be used to investigate sedimentation and seepage in rivers and dams, and neutron activation analysis will be used in Romania in developing methods for determining the purity of nuclear materials.

The use of radioisotopes in industry will undoubtedly be furthered as a result of electronics and instrumentation training courses and development work in Turkey, Burma, India, and Argentina, and gamma radiography will be developed in Thailand and Indonesia. A project on radiation treatment of plastics to increase their strength will be pursued in Yugoslavia, while in Colombia, processing of plastics, wood, and pharmaceuticals with ^{60}Co will be extended, and in Hungary a dosimetry system for use in radiation processing of pharmaceuticals will be developed.

The use of isotopes in medical diagnosis will be promoted in Sofia, Bulgaria, and at the Korle By Hospital in Ghana. At the Avicennes Hospital in Rabat, Morocco, emphasis will be on radioisotope use in diagnosing blood diseases and in Rangoon, Burma, on blood diseases and kidney function. In Israel, the relations between coronary heart disease and race will be studied with radioisotopes.

Agricultural applications of isotopes will be investigated in Vietnam and Iran, the work in Iran being an extension of some already under way. Radiation-induced mutations in wheat and barley will be studied at agricultural experiment stations in Lebanon, in Turkey, and at Giza, UAR. The possibility of eradication of the Mediterranean fruit fly by releasing radiation-sterilized males will be investigated in Cyprus,

and food preservation by irradiation will be studied in Thailand and India. Radioisotope tracer techniques will be used to study plant physiology in Cambodia, in attempts to increase rubber-plant yields, and to study fertilizer efficiency in Uruguay. Phosphorus in Greek soils will be studied by radioisotope techniques, as will the agricultural use of water in arid regions in Chile.

(RHL and MG)

Recent Accessions of Isotopes Information Center

Conferences

Advances in Energy Conversion Engineering, Papers, Critiques, and Summaries presented at Intersociety Energy Conversion Engineering Conference, Miami Beach, Florida, August 13–17, 1967, The American Society of Mechanical Engineers, New York, 1967. (CONF-670806) ($28.50, 1374 pp.)

Isotopes Studies on the Nitrogen Chain, Symposium Proceedings, Vienna, 1967, IAEA,* Vienna, 1968. (STI/PUB/161) ($7.00, 343 pp.)

Microbiological Problems in Food Preservation by Irradiation, Panel Proceedings, Vienna, 1966, IAEA, Vienna, 1967. (STI/PUB/168)($3.00, 148 pp.)

Nuclear Activation Techniques in the Life Sciences, Symposium Proceedings, Amsterdam, 1967, IAEA, Vienna, 1967. (STI/PUB/155) ($15.50, 709 pp.)

Preservation of Fruit and Vegetables by Radiation, Panel Proceedings, Vienna, 1966, IAEA, Vienna, 1968. (STI/PUB/149) ($3.00, 152 pp.)

Isotopes Information Center Reports

R. Curl (Comp.), List of AEC Radioisotope Customers with Summary of Radioisotope Shipments, FY 1967, USAEC Report ORNL-IIC-13, Oak Ridge National Laboratory, March 1968. (CFSTI,† 37 pp.)

M. Gerrard and P. S. Baker, Selected Abstracts of Non-U. S. Literature on Production and Industrial Uses of Radioisotopes (From *Nucl. Sci. Abstr.,* July–December 1967), USAEC Report ORNL-TM-1940 (Pt. 2), Oak Ridge National Laboratory, February 1968. (CFSTI, 59 pp.)

Division of Isotopes Development Reports

J. A. Kent, G. B. Taylor, W. R. Boyle, and A. W. Winston, Radiation Induced Polymerization of Vinyl Monomers Impregnated in Wood, USAEC Report ORO-2945-8, West Virginia University. Presented at the Winter Meeting of the A.I.Ch.E., Detroit, Dec. 8, 1966. (CONF-661201-3) (CFSTI, 17 pp.)

W. E. Loos and J. A. Kent, Fastening of Wood–Plastic Combinations, Topical Report, USAEC Report ORO-2945-9, West Virginia University, Sept. 1, 1967. (CFSTI, 38 pp.)

T. C. Martin, K. R. Blake, I. L. Morgan, C. V. Parker, Jr., and R. Sieberg, An On-Line Nuclear Analysis Facility, Interim Technical Report No. 3, USAEC Report ORO-2980-12, Texas Nuclear Corporation, June 15, 1967. (CFSTI, 56 pp.)

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A. F. Novak and J. A. Liuzzo, Radiation Pasteurization of Gulf Shellfish, Final Summary Report for the Period January–December 1966, USAEC Report ORO-654, Louisiana State University, Jan. 15, 1967. (CFSTI, 24 pp.)

J. R. Rhodes, T. Furuta, and J. D. Hall, X-Ray Analysis Using Radioisotope Sources, Interim Technical Report No. 3, April 1, 1966–May 31, 1967, USAEC Report ORO-3224-12, Texas Nuclear Corporation, July 14, 1967. (CFSTI, 101 pp.)

L. J. Ronsivalli, J. H. Carver, J. D. Kaylor, J. B. Huff, and E. J. Murphy, Study of Irradiated-Pasteurized Fishery Products and Operational Guide for MPDI, Period Covered: October 1, 1965–September 30, 1966, USAEC Report TID-24016, Bureau of Commercial Fisheries, October 1967. (CFSTI, 172 pp.)

*International Atomic Energy Agency (IAEA) publications may be obtained from the National Agency for International Publications, Inc., 319 East 34th Street, New York, N. Y. 10016.

†All CFSTI reports are available for $3.00 (full size) and $0.65 (microfiche) from Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Va. 22151.

C. P. Ross, C. L. Angerman, and F.D.R. King, Development of ^{60}Co Capsules for Heat Sources, USAEC Report DP-1096, Savannah River Laboratory, June 1967. (CFSTI, 47 pp.)

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D. E. Wood, Development of Fast Neutron Activation Analysis for Liquid Loop Systems, USAEC Report TID-24029, Division of Kaman Corporation, Kaman Nuclear, July 1967. [KN-67-458 (R)] (CFSTI, 31 pp.)

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J. Askill (Comp.), A Bibliography on Tracer Diffusion in Metals: Part V. Self and Impurity Diffusion in Simple Metal Oxides, USAEC Report ORNL-3795 (Pt. V), Oak Ridge National Laboratory, January 1968. (CFSTI, 11 pp.)

C. A. Baker, A Review of Gamma Activation Analysis, British Report AERE-R-5265, January 1967. (HMSO,* 20 pp.)

J. H. Clausen, Food Irradiation Activities Throughout the World, Business and Defense Services Administration of the U. S. Department of Commerce, 1968. (Superintendent of Documents, U. S. Government Printing Office, Washington, 20402, $0.50, 91 pp.)

C. G. Clayton, A Survey of the Application of Radiation Techniques in Oil and Mineral Boreholes, British Report AERE-R-5368, February 1967. (HMSO, 74 pp.)

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H. P. Yule, Instrumental Reactor Neutron Activation-Analysis Studies on 72 Elements in a Refined Hydrocarbon, *Nucl. Appl.*, **3(10)**: 637-640 (1967).

FIRST INTERNATIONAL CONFERENCE ON FORENSIC ACTIVATION ANALYSIS

The *Proceedings of the 1st International Conference on Forensic Activation Analysis,* held on Sept. 19–21, 1966, in San Diego, Calif., is available from Gulf General Atomic, P. O. Box 608, San Diego, Calif. 92112, for $7.00. The proceedings, edited by V. P. Guinn, who is well known for his pioneering work in forensic activation analysis, provide an authoritative and up-to-date state-of-the-art report on activation analysis and its use in forensics. The conference was sponsored by the Division of Isotopes Development of the U. S. Atomic Energy Commission, the California Association of Criminalists, and Gulf General Atomic. (FJM)

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